Appendix I

Comments Received from the University of California Peer Reviewers Regarding the CARB Draft Final Report

Section 57004 of the California Health & Safety Code requires external peer review of the scientific basis of prospective environmental rules and regulations. Although the Lake Tahoe Atmospheric Deposition Study itself would not directly result in any rule or regulation, the results of LTADS and subsequent water clarity modeling will guide the development of rules and regulations to address the declining water clarity problem. Because atmospheric deposition is a complex phenomenon, staff pursued external peer review via the Cal/EPA Interagency Master Agreement with the University of California, Office of the President (IMA 98-004). The task orders included review of the study plan, interim products, and the draft final report, titled "Lake Tahoe Atmospheric Deposition Study".

The Peer Reviewers selected by the University of California, Office of the President, were received from Professors Thomas Cahill (UC Davis), Keith Stolzenbach (UC Los Angeles), Gail Tonnesen (UC Riverside), Akula Venkatram (UC Riverside), and Tony Wexler (UC Davis). Each reviewer has expertise directly pertinent to atmospheric deposition. CARB staff is grateful for their insights, comments, suggestions, and desire to assist staff in conducting the study in a sound, technical manner. Their interest and review efforts, which greatly exceeded the available compensation, demonstrated their commitment for a solid, scientific foundation for the LTADS report and results. Where feasible and practical within budgetary and time constraints, staff has incorporated their input. The comments of the peer reviewers are presented in their entirety here in Appendix I. The major comments of the peer reviewers are repeated with the staff responses interspersed in Appendix J.

Comments Received from Professor Thomas Cahill

Peer Review of Lake Tahoe Atmospheric Deposition Study (LTADS) Draft Final Report of September, 2005

By Thomas A. Cahill Professor of Physics (Recalled) and Atmospheric Sciences (Emeritus) Head, UC Davis Air Quality Group 1970 – 1997, and Head, DELTA Group, 1997 - present

September 19, 2005 Revised October 12, 2005

Summary of Comments:

Overall comment: LTADS, by far the largest air quality study ever mounted at Lake Tahoe, represents a major enhancement of our knowledge of Lake Tahoe air quality and a major advance in our understanding of deposition phenomena. Because of its size and scope, it should be able to stand alone as a primary source for present and future planning and the basis for additional research. As it is written, it does not reach the stature I would have expected from such a massive effort. I recommend that additional funds and about a year should be taken to merge this study with prior and concurrent data.

Specific Comments:

Urgent: Error in the Primary Data:

While the DRI XRF and UCD S-XRF data are in excellent agreement for major elements before particle size self absorption corrections are applied, (example: 0.99 ± 0.06 , $r^2 = 0.94$ for silicon), the phosphorus data used by LTADS, when corrected for self absorption of the phosphorus x-rays in a soil matrix, are way too low by a mean weighted factor of times 7.7. Details can be found in comments on Section 3, but in summary:

- a. Based on the more sensitive S-XRF data and revised DRI MDLs, there are no statistically significant DRI phosphorus data.
 - i DRI claimed 26 (out of 70) positive values, with the average value/MDL
 - = 0.68
 - ii. Only 6 of these values had values > MDL, i.e., value/MDL > 1.0; 5 TSP_{10-35} , $1 \text{ PM}_{10-2.5}$,
 - iii Based on UCD S-XRF analysis, none of these was above the true MDL,

iv. DRI used $PM_{10-2.5}$ corrections for TSP_{10-35} samples (DRI e-mail, 9/2005). This gives a factor of roughly times 3 (+ 0.7 to + 2.4, or times 1.7 versus times 3.4) using DRI's self absorption estimates, which I show below are incorrect in any case for phosphorus in a soil matrix.

- b. UCD S-XRF provides the only valid phosphorus data. UCD saw 55 (out of 70 samples) positive values, with the average value/MDL = 2.6
 - i. 35 TSP_{10 35} above MDL
 - ii. 7 PM_{10-2.5} above MDL
 - iii. 13 PM_{2.5} above MDL
- c. However, neither UCD nor DRI used correct self absorption particle size corrections for phosphorus in a soil matrix (see Section 3 for the details). UCD S-XRF used only the negligible PM_{2.5} corrections needed for the DRI-UCD intercomparison. Thus, the UCD S-XRF phosphorus data must be multiplied by
 - i. times 8.3 for the 35 TSP₁₀₋₃₅ values (presumed mode at 15 μ m), since UCD used only the negligible PM_{2.5} corrections used in the DRI XRF UCD S-XRF intercomparisons.
 - ii. **times 5.0 for the 7 PM**_{10 2.5} **values** since UCD used only the negligible $PM_{2.5}$ corrections used in the DRI XRF UCD S-XRF intercomparisons.
 - iii. Times 1.0 for the 13 PM_{2.5} values.

Weighting these values by the observed data for particles > 2.5 μ m, one gets times 7.7 for the mean correction factor for coarse particles.

If the mode of the 35 TSP₁₀₋₃₅ values is larger than the presumed 15 μ m diameter, the correction will be larger (times 10.8 if the mode is 18 μ m, for example).

d. This will raise both the phosphorus dry deposition numbers and the phosphorus MDLs by a large number, as all agree that most of the phosphorus comes from large soil particles.

Comment #1: Only limited efforts were made to compare the year of the LTADS study to prior years at Lake Tahoe to put the study into historical context. Such a context is essential.

Comment #2: ARB and its contractors did not access or even list an enormous body of useful prior work at Lake Tahoe, including early ARB and US EPA work, TRPA sampling 1988- present, including archived IMPROVE samples and the much of existing data from ARB site at Sandy Way. Some of this prior work conflicts with the present studies. The net result is to greatly enhance LTADS uncertainties.

Comment #3: Uncertainties, especially for species like phosphorus, should be broken out into the specific categories before being combined into a single number. This

should include uncertainties in ambient measurements (siting, sampling and analytical uncertainties, year to year variability, sources not considered, ...) and deposition modeling (averaging and extrapolations, surface roughness and water deposition factors, inversions,...), perhaps including examples of prior efforts at deposition estimates..

Comment #4: The use of an upper cut point (TSP?) for the TWS eliminates particles and may eliminate coarse dust from roadways and strong wind events, thus underestimating phosphorus and soil dry deposition values.

Comment #5: TWS samplers delivered so little mass available for XRF analysis that even with good sensitivities, relatively few phosphorus data were observed during the entire study. The confusion appears to have involved the distinction between total mass collected (which was similar to IMPROVE) and mass aereal density in g/cm², the key parameters for elemental sensitivity by XRF (or S-XRF). The resulting high MDLs, even when alleviated by DELTA Group highly sensitive synchrotron-XRF, result in large uncertainties in the ambient data.

Comment #6: LTADS did not make use of prior and concurrent data on fine particles and phosphorus, including prior ARB programs, current TRPA programs, including over 5,000 values phosphorus values at South Lake Tahoe.

Comment #7: LTADS underestimates fine particle phosphorus input to Lake Tahoe because of its inability to see the low ambient levels (few ng/m3) involved and lack of source information on the automotive and diesel very fine phosphorus emissions. These emissions, plus wood smoke, are trapped under a shallow inversion at night and move out over a goodly fraction of the lake surface, enhancing deposition. This will cause LTADS to underestimate fine particle phosphorus deposition to the lake.

Comment #8: LTADS upper size cut aerosol capture still does not match the very coarse particle capture in the TRG deposition buckets. With the evidence of LTADS (and elsewhere) that there is significant soil (and thus phosphorus) mass above TSP limits, this will cause LTADS to underestimate lake deposition of very coarse phosphorus and soil.

Comment #9: Study 1.6.5 "...local generation of photochemical smog appears to be the main cause of increased O_3 within the basin." 25 years of studies, including some in LTADS, find that almost all O_3 within the basin is transported in from the west slope (see USFS Watershed 2000, fff). This comment is repeated in LTADS elsewhere. I have provided details of the prior arguments below in the appropriate section. The final report must either correct or support this statement, including evaluation of earlier studies, as otherwise this result would have a massive impact on TRPA in-basin regulations. (see comments section 8.10)

Overview of the review

I view my most important task is to make sure that any errors in the LTADS report are brought to the attention of the authors and thus corrected before the results become public.

I view my next most important task is to insure that the uncertainties associated with the results are appropriate, a difficult task in any case but especially difficult in the case of LTADS as the landscape is littered with the wrecks of worthy efforts to tackle the very difficult problem of atmospheric deposition.

My third most important task is to do what I can to place LTADS into a long term framework so as to insure that it can be rationally compared with prior work, typical metrological conditions, and long term trends, as any short term study, no matter how intensive, is subject to annual variations in sources and meteorology.

This being done, I must be realistic in that I am reviewing the report as it exists today, with relatively little chance to make any major changes in such a large effort with so many investigators and contractors. There is little to be gained in going over in any detail the original laudable, and probably unrealistic, goals of LTADS (the report itself on numerous occasions refers to initial expectations not met for a variety of reasons). There is little to be gained in reviewing suggestions I (and others) made in the original review of the LTADS design, many of which were not enacted for a variety of reasons. Whatever the original expectations, LTADS is a very important study, by far the largest air quality study of any kind ever undertaken in the Lake Tahoe basin. As such, it must stand tall as the foundation for all future studies and the basis for all future regulatory actions. Any who picks it up must get a whole overview of the past 35 years of air quality work, since the aerosol ambient concentrations are the key parameter needed to estimate deposition values. But one of the failings of LTADS is, that by using staff and contractors many of whom had little or no experience at Lake Tahoe, much prior useful information was not cited. In fact, the work was probably not even known, as much of the research at Lake Tahoe is in the form of reports as required by the constant regulatory focus, but not the refereed literature.

As a first connection to the past work, a complete bibliography is an excellent beginning. There are two major integrative peer reviewed studies that can provide much of this information – the 1996 USDA Sierra Nevada Ecosystem Project (4 volumes, but the three key papers are "Air Quality in the Sierra Nevada", "The Case Study of Lake Tahoe", and "Biological Effects of Air Quality") plus the USFS Watershed Assessment Study, 2000 (with the key paper Chapter 3, Air Quality). I have included the references from these papers in electronic form as a start to this process.

I have on occasions included plots and data from these last two summary papers since they were both exhaustively refereed. These data ware designed to focus, and occasionally challenge, the interpretations of LTADS, and help put it into a more global perspective. They are not intended to be inserted into the report.

Finally, I have to be honest in stating that this was a very difficult report to review. First, it is very large, and time is short. Second, much of the information is in individual reports, some of which are hard to get or not yet posted on the web site. Third, "the devil is in the details" that are often obscured in the relatively short summaries in the LTADS report. Finally, the areas of expertise required are so extensive that there are large areas that I can not review intelligently. In these cases, I will pose questions and hope that then experts in these areas address the problems.

Executive Summary

This needs to be re-evaluated after important corrections (circa x 3) are made to the predicted TSP phosphorus levels, MDLs, and better information on increased transfer rates from land to water. (see vii) Both directly affect phosphorus deposition estimates.

Page v "the most comprehensive database of atmospheric data ever assembled for the Lake Tahoe region." This is true only for certain parameters, and is not true for aerosol size and composition, in which LTADS is less than 1% of the existing aerosol data. For example, there is a more complete aerosol profile in (ARB 1979) at 10 sites within the basin, summer and winter. For parameters like soils, these data are still useful. Beginning in 1988, paired sites at Bliss SP (which measures only transported aerosols) and South Lake Tahoe (transported plus local aerosols) are available twice/week. Very limited use was made of these data. Recent TRPA studies included highly size and time resolved samples, including over 5,000 measurements of phosphorus.

1.1 History

Comment 1.1 – 1 In general, the report would be far more readable if the references were better cited in the text rather than using "passive voice" sentences and a terse reference. In paragraph 2, for example, you could state "Measurements made by Prof. Charles Goldman and the UC Davis Tahoe Research (TRG) Group (http://trg.ucdavis.edu) showed that between the mid 1960s and....." That way readers can go immediately to primary sources.

Comment 1.1- 2 LTADS would be well served to state at this point that it was designed to handle the most intractable but most important problem in the contribution of air quality to the clarity of Lake Tahoe, atmospheric deposition, but it was building on 35 years of air quality research in the basin, some of which, including the pivotal 1974 "Lake Tahoe Air Quality", was ARB funded.

Comment 1.1 – 3 Any short term intensive study must be put into the perspective of long term conditions at the site and nearby areas including source and meteorological changes that could reflect LTDA in the long term environment. For example, there was a major forest fire in Oregon in 2002 that lasted for a month and had a significant phosphorus contribution at Lake Tahoe not seen in typical low temperature Tahoe wood smokes, also tracked by non-soil potassium in the 0.34 to 0.56 µm size mode.

- 1.10 An important discussion, some of which should probably go into the Executive Summary, as it gives a sense of the uncertainties involved.
- 1.11 "Earlier analytical...." References needed to the 45 years of TRG work. Recent work (Schladow 2004 see http://trg.ucdavis.edu) shows that fine insoluble particles are a key factor in lake clarity through much of the year.
- 1.17 Deposition method comparison. Excellent. I have long recommended such a study.

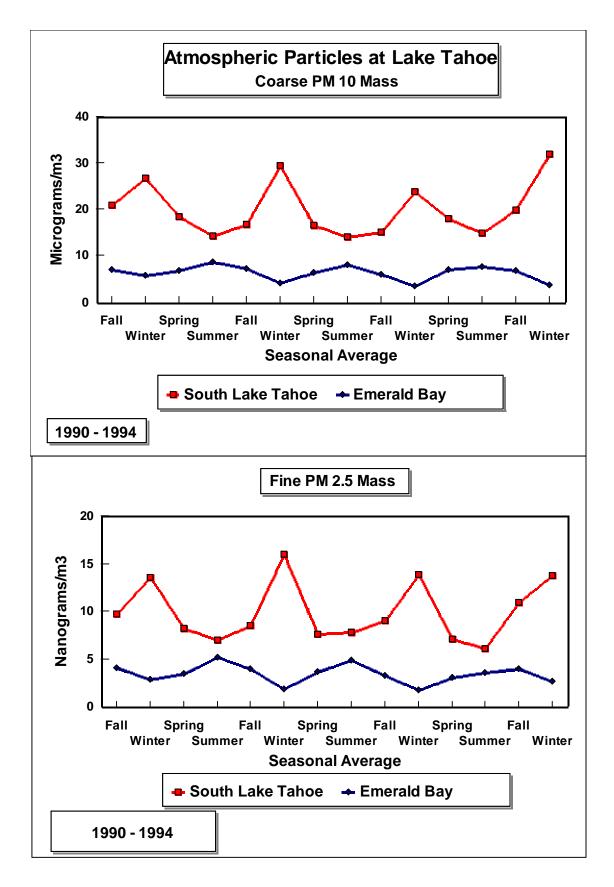
Special studies: These are important and useful, but all are of relatively short duration and at limited sites. Major extrapolations in space and time are necessary to fit these data into deposition models. These uncertainties are great and must be stressed.

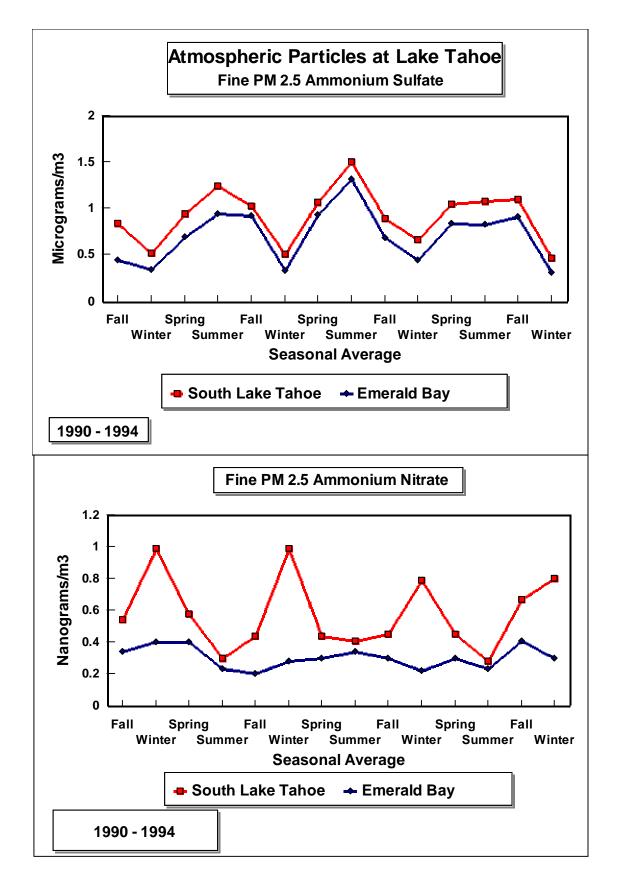
- 1.18 I deeply regret that the Thunderbird Lodge deposition samplers were deleted. We model important summer dust transport up Highway 89 and then across the lake.
- 1.23 HNO₃ "...urban areas to the west of Lake Tahoe can not be identified as an important source." This result contradicts airborne results of Carroll et al 1998 on the existence of perched layers of reactive species and ozone above the Sierra, some of which come down at night (also seen at Yosemite 2002)
- 1.24 Study 1.6.5 "...local generation of photochemical smog appears to be the main cause of increased O_3" I hope this was a typographic error, and not the conclusion of this report. 25 years of studies, including some in LTADS, find that almost all O_3 within the basin is transported in from the west slope (see USFS Watershed 2000, fff). The final report must correct this error and include an evaluation of the earlier studies. This is a good example of lack of coordination in LTADS and its contractors with potentially important results.
- 1.25 Study 1.6.9. This is one more example of how the delays in LTADS make these results of limited value to TRPA and Lahontan in its decision making process.
- 1.25 Study 1.6.10 This effort was very helpful to LTADS as the only phosphorus measurements > MDL made in the entire LTADS. I developed this system in 1997 based on earlier work at Stanford SSRL (Kuwait, 1991 ff), and Dr. Cliff has done a splendid job running and upgrading the system. We rely on it for all DELTA Group research projects, including the > 5,000 phosphorus values seen in the TRPA data (2002) but not considered for LTADS.
- 1.27 A vital study of deposition.

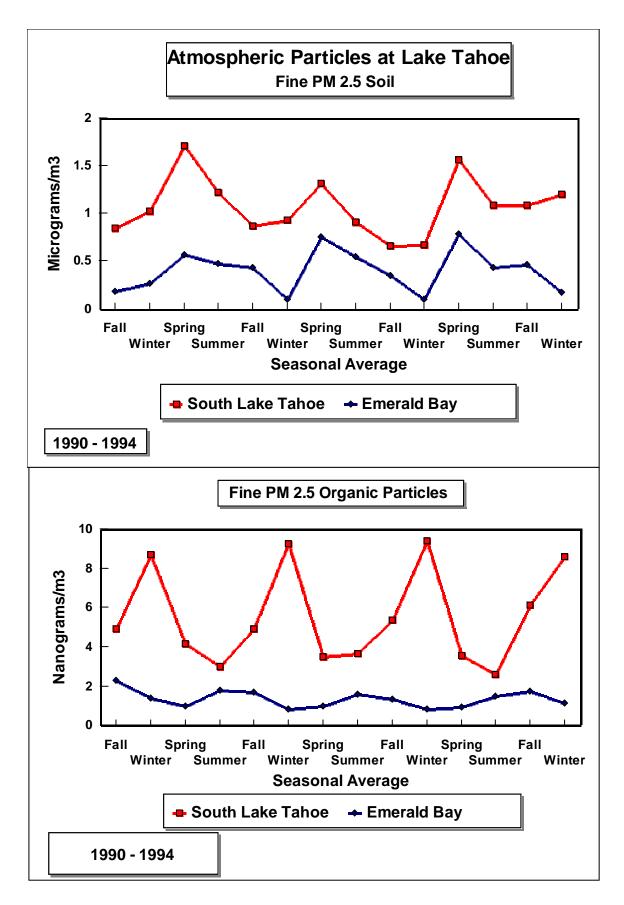
At the very least, a bibliography of prior work should be included. I have attached references from the USDA 1997 Sierra Nevada Ecosystem Study, Chapter, 49, "Air Quality in the Sierra Nevada". (Cahill, Campbell, Carroll, and Gill), and the USDA/USFS

Lake Tahoe Watershed Assessment Project (2000), Chapter 3, Air Quality (Cahill and Cliff) as a start.

As an example of earlier results with direct applicability to LTADS, I present below the USFS Watershed 2000 estimates of transport into the Lake Tahoe basin. These were based on the paired Bliss SP (BLIS) and South Lake Tahoe (SOLA) samplers, 1988- present. This enormous data set designed mostly for visibility studies was able to cleanly resolve transported samples (seen at BLIS) from local plus transported aerosols, (seen at SOLA). BLIS in fact in summer reflects exactly the Desolation Wilderness Area at Lake Aloha (USFS, 1992). The first plot shows ammonium nitrate, which is always a transported aerosol, and ammonium nitrate, transported in summer, local in winter.

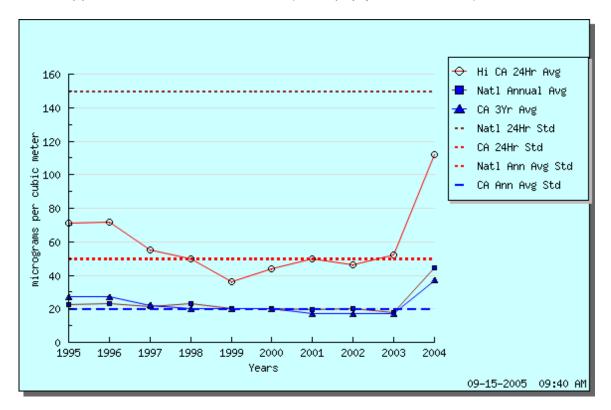


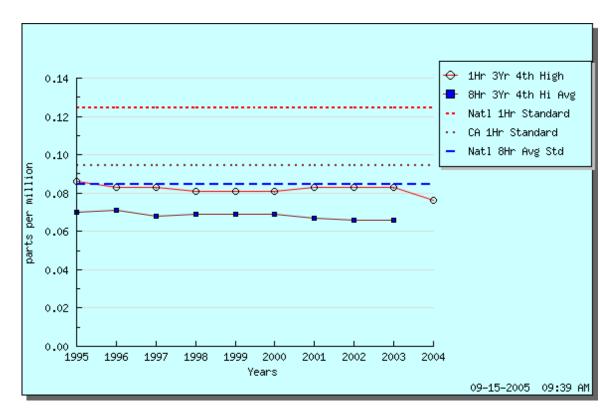




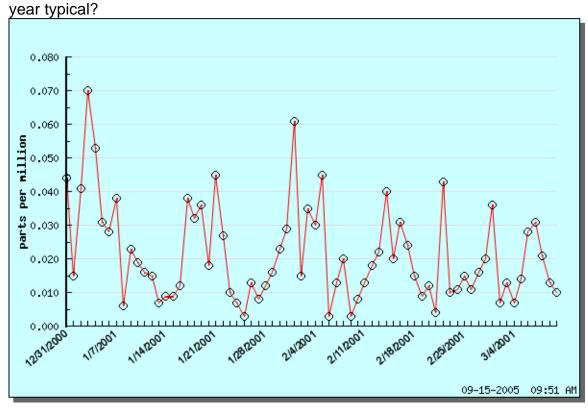
It is also easy to add trend data from the Sandy Way site, easy to do considering the availability of the splendid ARB ADAM resource, and meteorology from programs such as www.weatherundergound.com.

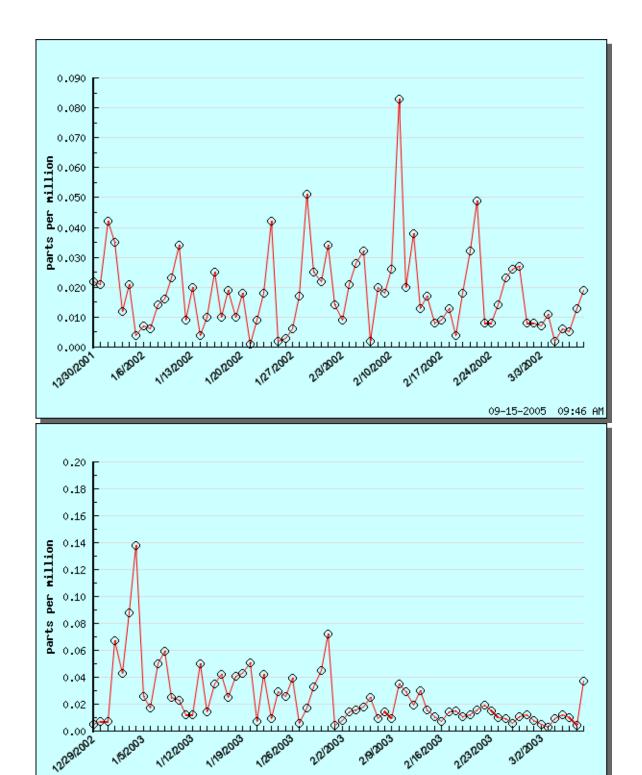
Examples of the ADAM trend data are shown below for ozone and PM_{10} . LTADS appeared to have taken place in a stable period. Note that the PM_{10} problem in Spring, 2004, appears to be a technical error. (D. Popejoy, ARB, 9/2005)



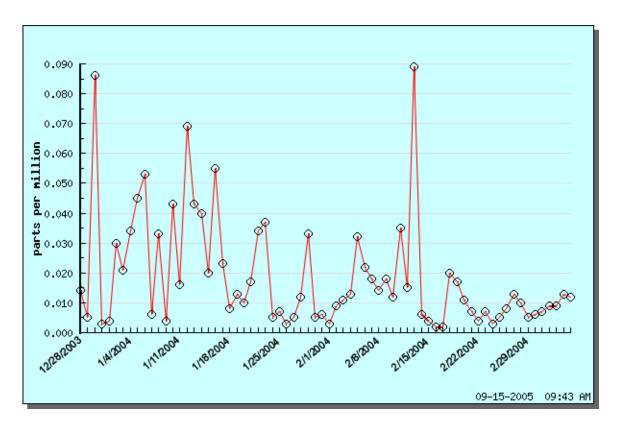


I have also shown the NO data for Sandy Way, 2001-2004, data that reflects mostly truck traffic as the source and meteorology as the dispersant. Note the important changes year to year and as the season progressed. To what degree was the LTADS





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Without these types of results, LTADS can not be put into a long term perspective.

2. Atmospheric processes

Good work. A very useful section.

- 2.12 2.2.3.2 See the surface based in versions in ARB 1979 (bulk Richardson number data) and USFS Watershed 2002, with pictures. The latter clearly shows the inversion blocking the smoke from a controlled burn from reaching the lake surface.
- 2.15 Thank you for using our data (Barone 1979) on inversions, as it was the only study to go as low as 50 ft above the lake. (ARB 1979) and thus catch the massive 9° surface inversion.

3. Data Quality and Summary of Ambient Concentrations

Comment: Aerosol data were merged into the USFS Watershed Assessment Lake Tahoe Airshed model (LTAM), a gridded Eulerian model with over 1,000 cells incorporating meteorology, all prior ambient measurements, and traffic data. It might be instructive to compare LTAM model results to LTADS measurements.

3.2 Question. What data supports the upper cut point of the TWS and the Mini-vol?

3.3 The TWS sampler design was not well coordinated with the proposed DRI XRF analysis protocol. The TWS collected 26 m³ of air but (correct me if I am wrong) spread the sample over a 47 mm filter. These results in and areal density of circa 13 cm²/26 m³, or 0.50 cm²/m³. This is the number that the XRF data (in ng/cm²) must be multiplied by to get ambient concentrations in ng/m³. For comparison, IMPROVE uses 32 m³ or air on a 3.5 cm² filter, for a conversion constant of circa 0.11, or almost 5 times better for an equivalent XRF MDL. Finally, the DRUM samplers, with coated substrates and not limited by filter clogging limitations, has a conversion constant of 0.017 cm²/m³, or roughly 30 times better than the TWS. This is about the ratio seen at Lake Tahoe, 30 ng/m³ for TWS, 1 ng/m³ for the DRUM.

- 3.4 The DELTA Group and IMPROVE (when I ran it) have always had excellent agreement with the DRI XRF system.
- 3.18 We too use Bevington (1969) for our statistics.
- 3.20-3.22 Figure 3.3 The 50% difference between the sum of species and TSP mass for the entire LTADS data set, but best seen at SOLA and Sandy Way, may represent an inadequate correction for self absorption in the TSP XRF data. Note that because other species contribute to TSP (OC,...), and some soil species are heavier and have smaller corrections, (Ca...Fr...), the actual XRF corrections for light elements will be still larger. This especially affects the phosphorus data (see above) that may be low by a factor as much as 3.
- 3.52 The "TWS period" is confusing. In addition, there are periods that overlap in season, and if these are plotted in the same segment, we can get a better idea of year to year variability (see above). Note that there was an unusual forest fire in summer, 2002. (Just a comment. I will do this myself don't bother changing the report).
- 3.71 Section 3.2.4 These are very valuable data and, while not new (we saw the same in ARB, 1979, for example, but only at SOLA) they greatly increase our knowledge of local behavior. Good work.
- 3.91 This is a very important section, but would benefit by matching with meteorology and using CalLine 4 type modeling.
- 3.92 Good use of the Gertler/Tanay data. Very important results.
- 3. Data Quality and Summary of Ambient Concentrations
- 3.2.5 Phosphorus

Corrections to existing LTADS data

A great improvement was made to the LTADS phosphorus data base by reanalysis with the DELTA Group S-XRF system at the Advanced Light Source, Lawrence Berkeley NL. There are by now excellent and extensive data showing that S-XRF is both extremely sensitive and accurate. Below we summarize all DELTA Group S-XRF inter-comparisons in the past 5 years. Note that there were problems with the ARB RAAS analyses since the two internal ARB X-RF to ARB RAAS comparisons agreed only at the level 1.29 ± 0.63 for all co-measured elements. (DQAP v. 8.02, pg 32) We also give averages below without the ARB RAAS data.

Study and date	Methods	Average ratio, Al to Fe	Std. dev.	Average ratio, Cu to Pb	Std. dev.
BRAVO, 1999	PIXE vs S-XRF	0.99	0.04		
BRAVO, 1999	CNL XRF vs S-XRF			1.24	0.14
FACES, 2001	ARB XRF vs S-XRF	0.93	0.21	1.02	0.08
FACES, 2001	ARB RAAS vs S-XRF	(0.98)	0.27	(0.74)	0.23
ARB LTADS 2005	DRI XRF vs S-XRF	1.037	0.085	0.907	0.009
All prior studies	Average (no RAAS)	0.984 (0.985)	0.15 (0.11)	0.977 (1.055)	0.115 (0.076)

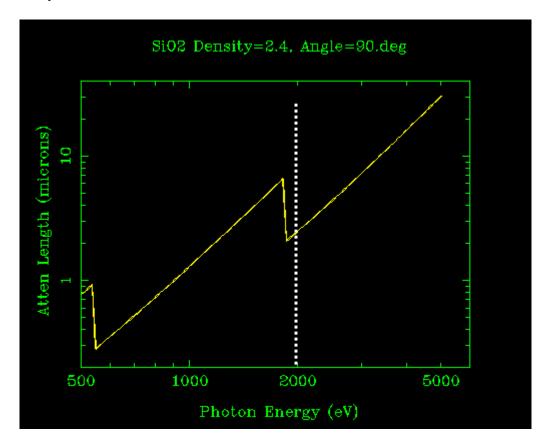
The excellent agreement for major and minor elements includes the LTADS/Cliff comparisons in LTADS. It should be noted that all these comparisons used PM_{2.5} filters, in which particle size corrections are small. The LTADS data, on the other hand, includes also PM₁₀ and TSP data. However, the data presented by DRI and UC Davis (Cliff, 2004, Appendix A) make no mention of particle size corrections for the phosphorus data. These corrections are required since some of the phosphorus (and other light element x-rays) are attenuated within the particle itself. These corrections can be estimated by using the mass extinction programs of the CXRO/Advanced Light Source LBNL. We calculated these corrections and had them reviewed as part of a different DRI study on diesel exhaust (B. Zielenska et al, 2002) using the same S-XRF system but a MOUDI impactor. These are presented in Appendix C.

The key point is that while such corrections are minor for fine particles (times 1.08 for typical $PM_{2.5}$ soils with an assumed mode at 1.8 μ m diameter), they become large for coarse and PM_{10} aerosols (1.3 for an assumed soil mode circa 5.6 μ m diameter, 1.8 for an assumed soil mode at 10 μ m diameter) and massive for TSP particles (times 3.4 for assumed soil mode circa 18 μ m diameter, but uncertain to \pm 30%)

However, there is a special problem when analyzing phosphorus in a soil matrix. The energy of the phosphorus x-ray is adequate to ionize aluminum and silicon atoms,

tripling the self absorption of the phosphorus x-rays. This is not a well known problem in ambient samples (but a massive problem in metallurgical analyses). Note that the matrix we typically use is based on diesel exhaust, and is thus irrelevant to phosphorus in soil.

As an example, below we show the transmission of x-rays through SiO₂, the largest single component of soil. The dotted line represents the energy of a phosphorus x-ray at 2.014 keV.

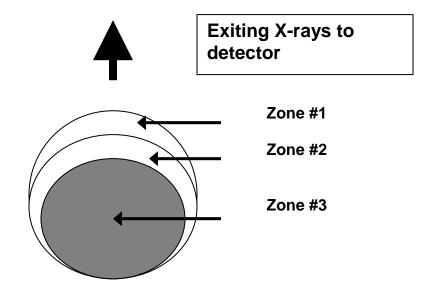


These mass attenuation data were complied from the Center for X-ray Optics (CXRO) web site at Lawrence Berkeley NL. http://www.cxro.lbl.gov

With these better attenuation coefficients, we must still handle the problem of the geometry of the larger particles.

Topic #1 Spherical geometry for self absorption corrections

The model I used is shown below. Zone #1 is the first soil layer (Al Si, K, Ca, and Fe plus typical oxides by the IMPROVE formula) in a spherical particle with radius r_o . Assume the x-rays are uniformly excited through the particle volume (we will modify this assumption later in Topic #2)



Zone #1 is the first ½ transmission layer, thickness $t_{1/2}$, which in our case is 2.15 µm thick. This means that x-rays at the back edge are attenuated by ½ reaching the front edge (and then free to travel to the detector), while x-rays generated near the front edge have no attenuation. Thus, ¾ of all these X-rays can reach the detector. The volume of this zone is the area, πr^2 times $t_{1/2}$.

Zone #2 has the same $\frac{3}{4}$ factor within the zone, but then has $\frac{1}{2}$ the x-rays removed in Zone #1, so that $\frac{3}{8}$ or 0.375 of the x-rays can reach the detector. The volume of this layer is slightly less than that of Zone #1 (we will assume equal for simplicity).

Zone #3 is now an ellipsoidal particle with a volume approximately $4/3 \pi$ ($r_o - \frac{1}{2}(2 t_{1/2}))^3$. A very small fraction of the x-rays in this Zone ever leave this zone, < 10% for a very coarse particle, and then they are attenuated by a factor of 4 passing through Zones #1 and #2. So the contribution of Zone #3 to the x-ray flux is only a few percent (set at 10%).

For the calculation we will assume the x-rays come from a volume of Zones #1 and Zone #2, 0.75 from Zone #1, 0.375 from Zone #2, summed to give 1.225 of the ideal thin sample yield of 2.0 if neither zone absorbed x-rays.

For an 18 μ m particle with r_o = 9 μ m, the volume of Zones #1 plus #2 is 0.245 of the volume of the entire particle, the remainder of which yields only 10% of all x-rays in an no-absorption situation. The correction factor is thus times 9.7, a significant reduction from the times 11 correction of a layered cube (9/27).

Topic #2 Beam attenuation

The x-rays can not be emitted unless the excitation reaches the atom. Picture the figure above with the exciting x-rays entering, and rotated 90o. Using an effective S-XRF excitation energy of 5.0 keV, this amounts to a factor 0.9 for an 18 μ m particle soil matrix, 0.8 at the back surface, 1.0 at the front surface. For a PM_{10-2.5} particle, the transmission factor is 0.96.

Summary:

The net correction for a spherical 18 μ m particle is thus 0.9 times 0.103 = 0.093, for a multiplicative correction factor of 10.8.

Thus, for the very large particles, the net correction to be applied to the DRI data is an additional factor of times 6.3 assuming and 18 μm very coarse particle mode, since it was already corrected by times 1.7, and for the UCD S-XRF data, times 10.8. Thus, for the very large particles, the net correction to be applied to the DRI data is an additional factor of times 4.9 assuming and 15 μm very coarse particle mode, since it was already corrected by times 1.7, and for the UCD S-XRF data, times 8.35. For PM_{10-2.5} mode particles, assume a mode at 7 μm , the net correction to be applied to the DRI data is an additional factor of times 3.0, since it was already corrected by times 1.7, and for the UCD S-XRF data, times 5.0. However, all the UCD S-XRF data provide the only statically sound phosphorus data to LTADS ambient values, so the corrections factors for all phosphorus data are:

- i. times 8.3 for the 35 TSP₁₀₋₃₅ values (presumed mode at 15 μ m), since UCD used only the negligible PM_{2.5} corrections used in the DRI XRF UCD S-XRF intercomparisons. For a presumed 18 μ m mode in the very coarse particles, this becomes times 10.8.
- ii. times 5.0 for the 7 $PM_{10-2.5}$ values since UCD used only the negligible $PM_{2.5}$ corrections used in the DRI XRF UCD S-XRF intercomparisons.
- iii. times 1.0 for the 10 $PM_{2.5}$ values.

Since all evidence (LTADS filters, IMPROVE, TRPA DRUM,) concur in the dominance of large particles in the ambient phosphorus concentrations, I propose that the ambient data must be corrected by these factors before the Final Report is released.

Potential resources for future work

"and drawing on other phosphorus data "

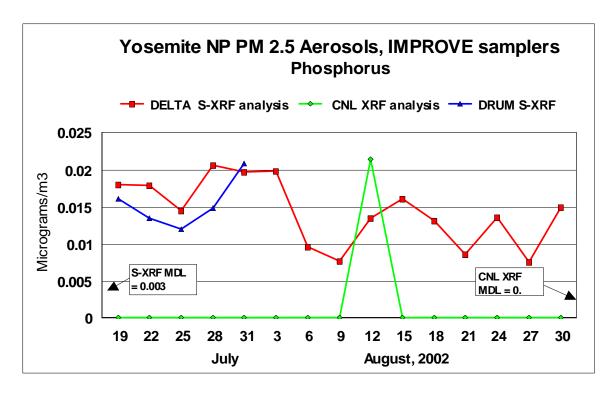
There are several other sources of phosphorus data that could have been utilized to reduce the phosphorus uncertainties. They should be at least listed so that future programs can have better success:

- 1. Archived filters capable of S-XRF analysis
 - a. The 6 months of week long integrated samples taken as part of the ARB funded UC Davis studies, 1978-1979 are available for analysis. They have

- been previously analyzed by PIXE (ARB 1978; ARB 1979) at 7 to 9 sites, summer and winter, but lacked sensitivity to see phosphorus using the current analytical technique. (Proton Induced X-Ray Emission) However, many other elements are available, some of which are summarized in the USFS Watershed Assessment Report, 2000 (Cliff and Cahill, Chapter 3). They are archived by the DELTA Group.
- b. DELTA Group removed the South Lake Tahoe samples from the IMPROVE sample archive, and thus we Group possesses hundreds of PM₁₀ and PM_{2.5} samples, 1989 2002. While the PM_{2.5} were analyzed by phosphorus (and the data summarized in this report) there were only circa 30 detects of phosphorus. The PM₁₀ filters, now known to have far more phosphorus than the PM_{2.5} samples, were never analyzed, and thus potentially could give hundreds of new data points on phosphorus.

2. TRPA DRUM data, 2001 – 2002

a. Over 5,000 measurements were made of phosphorus at South Lake Tahoe as part of a TRPA program on aerosols (Cahill et al, 2002). The samples were collected in the well characterized and theoretically well under stood Davis Rotating-drum Unit for Monitoring (DRUM) sampler (Marple et al, 1984; Rao, 1979; Raabe et al, 1989) while the analyses were done by the same S-XRF system used by LTADS to enhance their sensitivity (Cliff, 2004, Appendix A). The rotating drum technique was pioneered by the ARB, 1972 - 1977 (Feeney et al, 1975; Flocchini et al, 1976; Barone et al, 1978, Motellabi et al, 1992,....) and in its various enhancements has been used in over 30 studies around the world by ARB, IMPROVE, DOE, NSF, EPA, and NOAA, as well as many state organizations. The version used by TRPA was developed for EPA at BRAVO (1999) and enhanced by the ARB FACES program, 2001. The most recent evaluation was done by the NPS/IMPROVE at Yosemite, 2002 (see below).



These results make several points clear:

- 1. The IMPROVE filters, run specially for the study and analyzed by S-XRF, were similar to the DRUM data, despite the difficulty of having to add 144 individual DRUM analyses to male a single filter analysis.
- 2. The standard IMPROVE analyses at Yosemite, based on standard XRF at Crocker Nuclear Lab, clearly have much poorer sensitivity than either DRUM or filter S-XRF analysis. In addition, it appears that the quoted IMPROVE MDLs are unrealistic by about a factor of 10, a result also recently concluded by the IMPROVE leadership (White et al, AAAR, 2004). This affects the MDLs seen by IMPROVE in the PM_{2.5} filters at South Lake Tahoe (SOLA) and Bliss State Park (BLIS).

The TRPA DRUM data have several advantages that could have been used by LTADS:

- 1. Sensitivity -- MDLs are bout 10 to 20 times better than LTADS
- 2. Sizes -- 8 size modes are always collected, including a TSP equivalent to 35 $\,\mu m$ diameter
- 3. Time -- These data are available every 3 hours, and thus track the strong diurnal patterns seen in the BAMS data but with chemical information.

While it is too late to rectify these deficiencies in LTADS, the existence of these resources should be included as a guide to future work.

3.98 3.2.6 Dust experiments The new work (Schladow et al 2004) showing the importance of fine soils in lake clarity make dust data much more important.

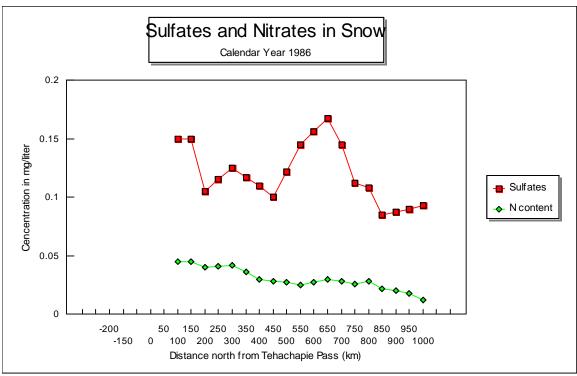
- 3.103 Reliance on the data of one day (March 12, 2004) on weak night winds without a lot of supporting data on weather, road conditions, and other factors are not very useful. We saw at the same site in different conditions massive transport from the road over the lake (ARB 1979).
- 3.107 How do these data compare to TRPA averages? Since there are clearly factors of 2 year to year, these directly affect the uncertainties in any deposition result based on aerosols.

4.0 Dry Atmospheric Deposition

4.48 This section needs work. The BAMS data and earlier (ARB 1979, USFS 2000) work show that the maximum of the aerosol, data occurs just as the winds are making the transition from weak downslope at night o strong upslope in daytime. The 6 to 8 PM period occurs in this latter condition. Note also that there is clearly aerosol mass even beyond 30 μm, data that are "iffy" with optical particle counters.

5.0 Wet Deposition

There is an important and unique deposition study by Laird et al (1984) cited in SNEP 1996. He measured nitrates and sulfates in snow from Canada to Mexico.



Note that BLIS is about 500 km north of the Tehachapie pass.

6.0 Air Pollution Transport

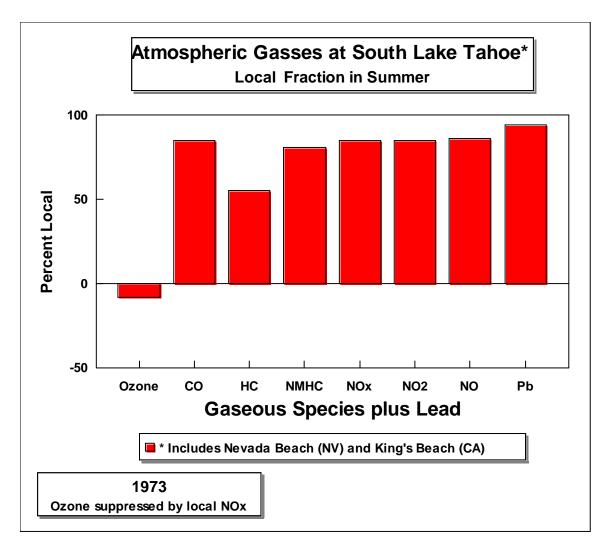
6.10 Section 6.6 Excellent summary that matches well prior work, including the low deposition velocity of the aged, transported plume and the blocking effect up and down of the prevalent surface inversions (USFS Watershed 2000).

6.25 Arrow #5 has to face the inversion problem (see above).

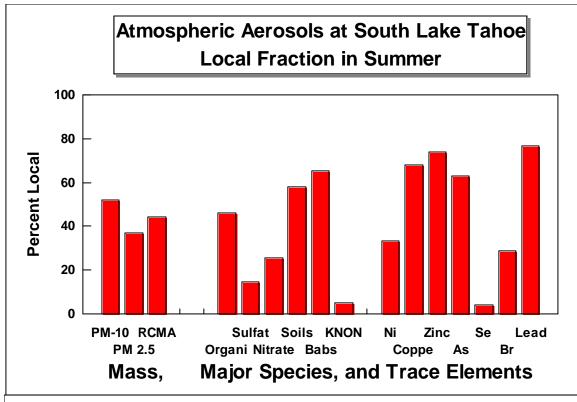
The addition of the Big Hill site and the data from it are a major enhancement of the LTADS program.

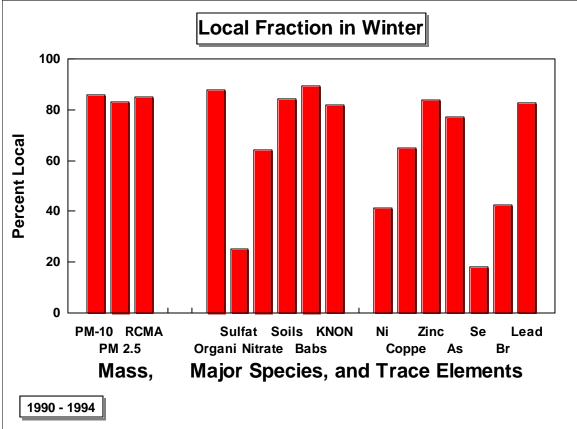
We do not concur with Carroll et al, 1998, "high concentrations of pollutants do not appear to reach the high altitude slopes of the Sierra" as shown by the results of Gertler et al, showing that for many parameters, including nitrogen, maxima rates of deposition are reached at the Sierra crest. We have long agreed that a sharp fall off of pollutants occurs just east of the crest (ARB 1978 ff).

Using the pioneering ARB report (1974), we can estimate local fractions in summer from spatial gradient data in the basin (summarized in the USFS Watershed Assessment (2000).



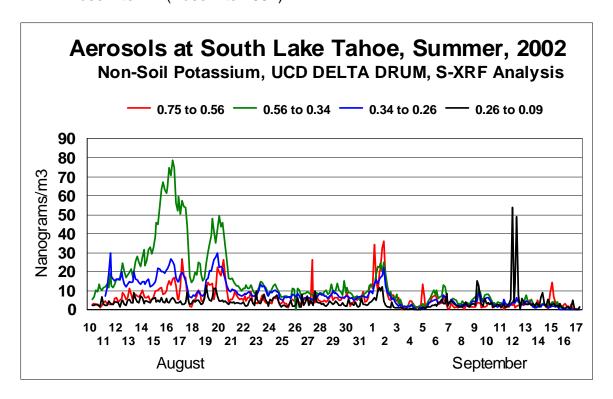
Using the decade of TRPA BLIS to SOLA comparisons, (above) we can estimate the local fraction of aerosols, summer and winter (USFS Watershed Assessment 2000)





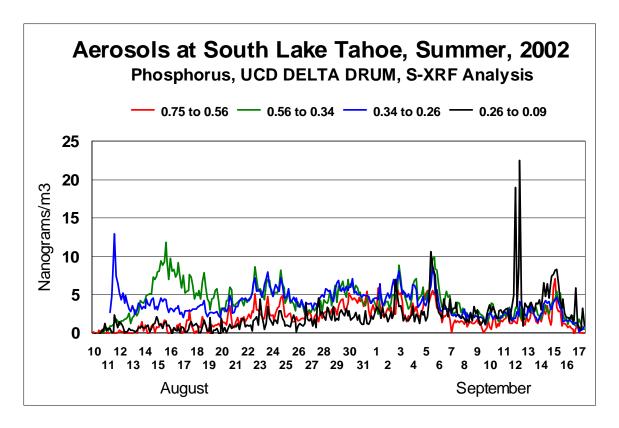
While we regret that the direct measurements made by Carroll et al under LTADS were not adequate to see phosphorus, we do have firm data on two forms of phosphorus transport into the Lake Tahoe basin:

- 1. Spring winds across the Sierra crest including
 - a. Local valley soils, (TRPA 1988 2002 in USFS 2000) and
 - b. Long scale transport of relatively phosphors rich Asia dusts (Seinfeld et al, 2004).
- 2. Phosphorus in forest fires, as seen in summer, 2002. (TRPA 2003) and at Yosemite NP (Yosemite 2004).



The phosphorus occurs at the same time and in the same size mode as the non-soil potassium, while NOAA HYSPLIT back trajectories track the air to southwest Oregon 4 days before (we also saw this at Yosemite NP). Vigorous updrafts can loft and transport abundant phosphorus flakes seen in coarse mode particles (Turn et al, 1997)

The importance of the summer, 2002 forest fire episode at Lake Tahoe was that TRG (Reuter et al, May, 2004) were able to compare the TRG phosphorus bucket data to the predictions of LTAM (USFS 2000) for this event with a reasonable good match. (circa 30%) This is the first instances that I know of where we have validated at least one portion of the deposition prediction, aerosols by size, time and composition versus measured deposition of phosphorus. LTAM was previously supported in its good match to the Spooner Summit controlled burn, 1999 (Cahill and Cliff, USFS 2000).



There is also some phosphate in local wood smoke, but it is closely tied to whether the coarser phosphate ash is resuspended in the smoke. This occurs in vigorously burning open fire places but not in the modern energy efficient wood stoves requires at Lake Tahoe.

Also note the phosphorus spikes September 12 and 13. This is from diesel and smoking car exhaust, tied to the use of zinc thiophosphate stabilizers in lubricating oil. This source is surprisingly important since it settles slowly as it is pushed by weak downslope winds each night over the entire lake area.

7.0 Characterization of PM and Nutrient (N & P) Sources

General Comment: While standard emission inventories at Lake Tahoe are weak (hence the DRI Road Dust and other studies) the situation with regards to phosphorus is nothing short of awful.

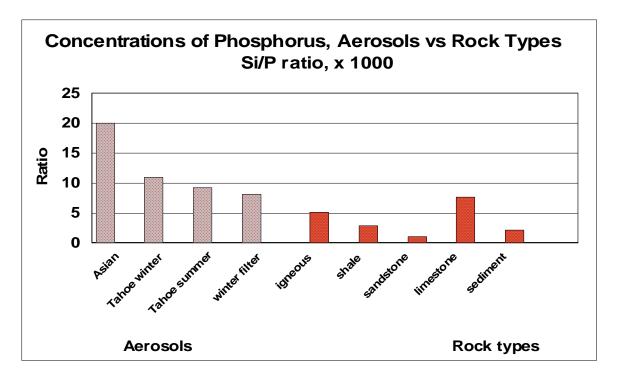
7.10 The reliance on a Sand Harbor, NV, site to characterize dust transport from roadways must be supported, as that location is characteristic of no other site in the basin (very narrow coastal plain, precipitous eastern mountain wall, low traffic volumes, little heavy truck traffic, unusual soil, little sanding and salting operations in winter, ,...). There are prior ARB data from other sites including South Lake Tahoe (ARB 1979) that show far more dispersion of road dust at that site, both in concentration and distance. This can be caused by a number of factors:

1. The z_o surface roughness parameter is high at SOLA because of the broken cover of roads and trees, This will loft and disperse pollutants,

- 2. Heavy truck traffic itself mixes the air to heights roughly 1.5 times the truck's height.
- 3. Heavier traffic and low wind velocities cause effluent heating in the waste heat of cars and trucks, which will maximize at dawn and dusk during rush hours. (ARB 1974 Freeway Study)

This is a key point because if one assumes a rapid removal of coarse particles, lake deposition values drop very sharply.

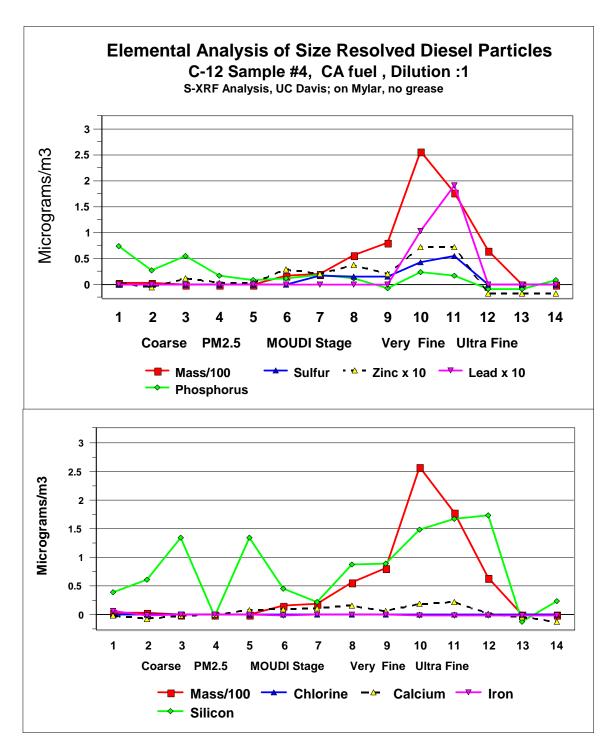
7.13 A second point is that the dust study "no detected phosphorus above uncertainties". Thus, we must assume that the estimates of aerosol emissions of phosphorus are based on the phosphorus content of soils. In fact, we have found at numerous sites that the phosphorus content of road derived aerosols is greater—than that of the original soil matrix (below). Thus, the phosphorus estmates will be about a factor of 2 too low based on road soils.



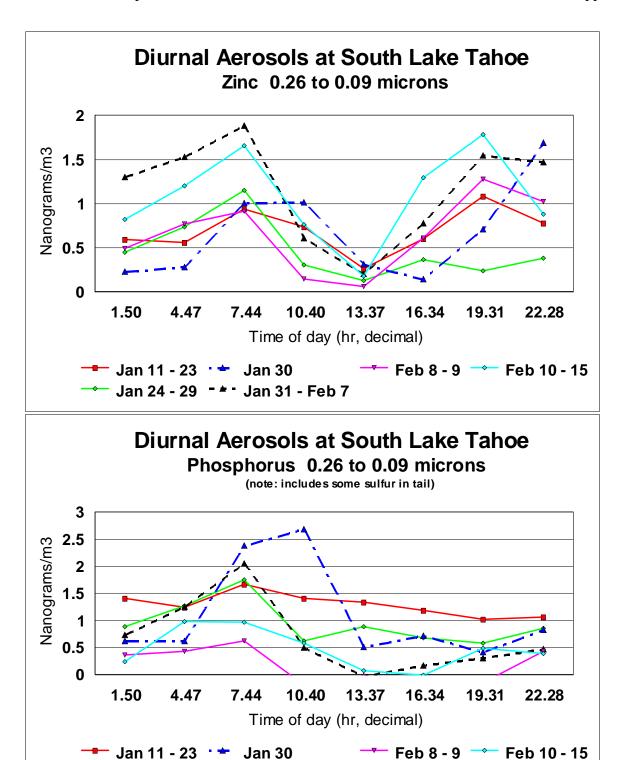
Note the high "Asian" phosphorus content of transported soils. These values were used in the Spring Asian dust transport with the frequencies of VanCuren and Cahill, 2002.

7.14 "One the other hand, phosphorus concentrations were so low as to be below the limits of detection." Recent data from DRI (Gertler et al, 2002, Zielenska et al, 2002) and the American Lung Association (Cahill et al, 2004) have tied truck/car exhaust to phosphorus emissions via the zinc thiophosphate in lubricating oil plus sulfur in diesel fuel.

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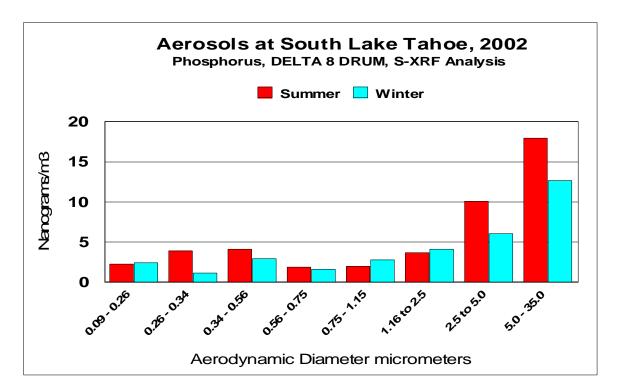


The very fine calcium comes from calcium carbonate, an ant-acid in the oil. This very fine (< $0.25 \, \mu m$) phosphorus and zinc are seen in the diesel spikes on Sept. 12, 2002 (above) and associated with traffic peaks in the very fine modes at the South Lake Tahoe SOLA site (below).



Finally, even in the average data, summer and winter, very fine phosphorus is seen.

Jan 24 - 29 - - Jan 31 - Feb 7

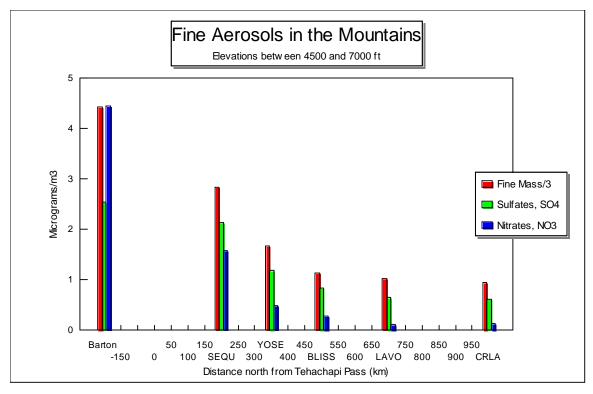


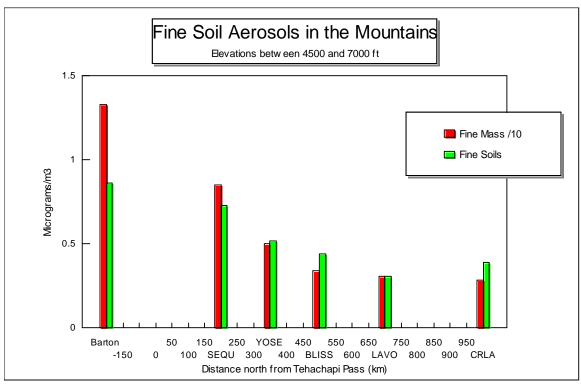
The particles from about 0.34 to 0.75 µm are from wood smoke, which in the summer was dominated by the Oregon fires (above)

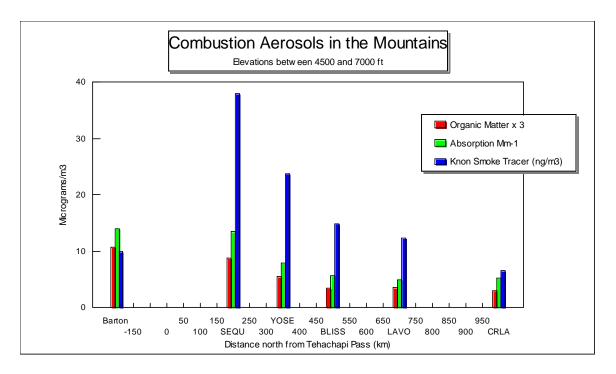
- 7-19 The phosphorus results of Turn et al 1997 in JGR give the phosphorus content of smoke. This should have been at least cited.
- 7-22 Section 7.3 ff Good work. It should be used to say to what extent the LTADS year was typical. These results would have been most useful much earlier in the report.

I have included similar 3 plots from Cahill et al (SNEP 1997) looking at two factors;

- a. The nitrogen and sulfur profiles, showing that Tahoe is a little north of the Bay Area influence,
- b. The soil profiles. Note how flat they are all the way to CRLA. We now know that that is due to Asian transport,
- c. Optical absorption, a tracer of smoke (no longer available from IMPROVE).



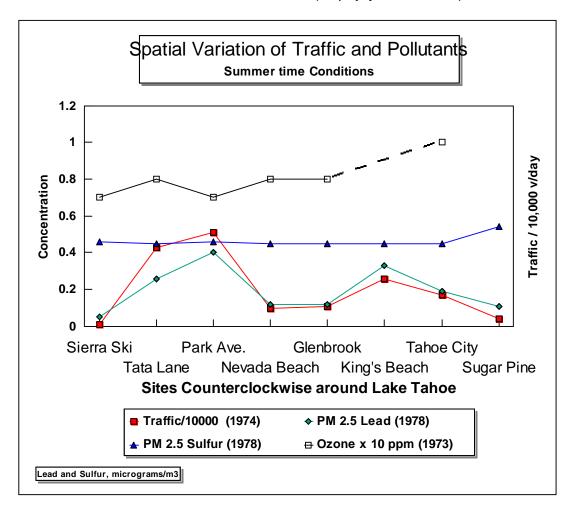




8-1 Conclusions, Lessons Learned, Insights, and Recommendations.

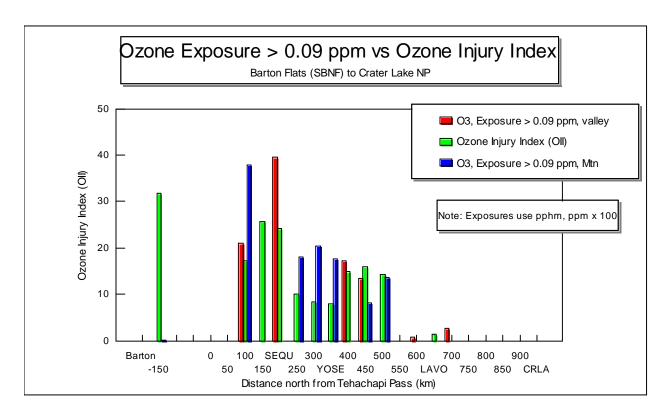
- 8.1 The Conclusions should be changed to match the revised Executive Summery, and reflect any corrections needed (viz, phosphorus ambient concentrations deposition rates,...).
- 8-3 ff A good section. Should be much earlier in the report.
- 8.2 Particulate matter. In winter, yes, roadways are dominant. In spring, roughly 50% of the soil is transported in (USFS Watershed 2000), and in summer, many sources are operational, not just roadways (TRPA 2002)
- 8.8 Phosphorus As I mentioned before, the phosphorus detection problem was compounded by a sampler design problem in the TWS. Our DELTA Group, UC Davis SXRF is at present the most sensitive non-destructive instrument in the world, which is why we developed it from 1992 to present and use it on DRUM and (occasionally) IMPROVE samples. It could be applied with great effect to the hundreds of archived PM_{10} samples from Lake Tahoe, 1978-2004.
- 8.9 Air pollution transport
- 8.10 "...ozone transport may occasionally occur." I believe abundant data show that essentially all the ozone at Lake Tahoe is transported, as shown by
 - a. O₃ peaks that occur after the sun has set,
 - b. uniform O₃ profiles across the basin when sources are localized,
 - c. the short residence time in the basin for reactions to occur,
 - d. the inversion barrier that keeps precursors at ground level,

e. 10 year temporal profiles that were slightly rising in response to increasing western Sierra (Highway 50) sources versus decreasing sources within the basin (Popejoy, ARB 1993)...



Either prove to me that ozone is regularly made in the basin or delete this comment.

Comment: The relatively lack of O_3 damage mimics the results of Petersen and Cahill (1988) and matches the predictions of Palmer and his Ozone Injury Index (OII) and Cahill et al (both in SNEP, 1997) on the damage threshold (below). Bliss SP is roughly 500 km north of Tehachapi pass.



8.11 The conclusion is correct and actually stronger than they know.

Appendix A

Urgent communication to ARB, 9/13/2005:

Leon

I am moving well on this report but I have come across a problem. I can not find out from the report whether (or what) self absorption corrections were applied to the phosphorus and soil data from DRI. I know they were not applied by Steve, who matched very well the DRI results for the major elements.

Could you contact DRI for me and inquire what were the self absorption corrections that were applied to the DRI data?

Incidentally, the corrections I used for phosphorus in the TRPA report were not negligible – for fine P, x 1.08, for PM10 P, x 1.5, and for TSP P, x 3.4. Everybody agrees that most of the P is in the coarsest particles so this could change the results profoundly. It also affects the MDLs by the same factor.

Tom Cahill

From DRI -

Hello Leon,

The correction factors are the same for both PM10 and TSP

A1 = 2.4

Si = 2.08

P = 1.72

C1 = 1.43

K = 1.28

Ca = 1.23

No correction for Sulfur.

The 25% uncertainty is reflected in the uncertainty that is reported for each element. The Kevex machines that were used are also 30 years old and the author of that paper is dead. These self absorption corrections are being looked at again, but that is the definitive reference at the moment. This comparison may be very valuable in checking how necessary these corrections are with newer instruments. My initial findings with my new XRF machine has indicated that they may not be needed (or at least not be as large), but the old Kevex machines did require them. Steve

Leon -

First, the corrections used by DRI for PM10 seem fine to me, and are close to what I would use myself depending on the size distribution, taking into account the coarser particles and the iron rich soils of Tahoe.

Second, I may have misunderstood Steve when he mentioned corrections. He may have been referring only to the TSP corrections, which were the same as PM10 for DRI, Thus, we would be back to "apples and apples". I will send this email to him to get this clarified.

Third, I do not understand DRI's comment on the detector system. The particle size corrections are a function of four things -

1. the size of the particle (assumed spherical, which is iffy for big particles but every one uses this assumption),

- 2. the energy of the exciting radiation (which may not efficiently penetrate into the center of a larger particle)
- 3. The composition of the particle, which gives the mass extinction as a function of x-ray energy, and
- 4. the energy of the exiting radiation, usually the biggest effect. Thus DRIs' comment can only refer to a change in the incident exciting x-rays, as that may change from instrument to instrument.

Fourth, there is no way that the self absorption corrections for a PM10 particle, with a mode around 7 or 8 microns, are suitable to a TSP particle. There is some flexibility depending on what is meant by TSP - I used a mode of 18 microns for my calculations, since I had a 35 micron inlet cut point, but the TWS used I believe 20 microns, giving a particle mode more like 15 microns. The assumption is a very steep coarse particle profile derived from the (dreaded)DRUM data at SOLA.

Since P was never seen in the PM2.5, and we all agree the coarse particles are a big deal for P, we have got to get this right.

I will keep working at this. Tom

Appendix B

Note: These corrections assume a diesel exhaust matrix. They are far too low for phosphorus in a soil matrix.

		MOUDI stage	MOUDI stage	MOUDI stage	MOUDI stage	MOUDI stage	MOUDI stage	MOUDI stage	MOUDI stage	MOUDI stage	MOUDI stage	MOUDI stage	MOUDI stage	MOUDI stage	MOUDI stage			
Element	Energy	1	2	3	4	5	6	7	8	9	10	11	12	13	14			
	(keV)	18	10	5.6	3.2	1.8	1.0	0.56	0.32	0.18	0.10	0.056	0.032	0.018	0.01			
Na	1.041	8.6	3.520	2.116	1.496	1.285	1.149	1.076	1.045	1.026	1.015	1.007	1.003	1.010	1.000			
Mg	1.254	8.0	3.334	2.030	1.464	1.272	1.149	1.084	1.055	1.032	1.018	1.009	1.003	1.010	1.000			
Al	1.487	6.2	2.732	1.745	1.341	1.203	1.112	1.063	1.041	1.023	1.012	1.006	1.003	1.010	1.000			
Si	1.740	4.0	1.989	1.391	1.184	1.111	1.062	1.036	1.023	1.014	1.009	1.004	1.002	1.001	1.000			
Р	2.015	3.4	1.787	1.292	1.138	1.082	1.044	1.023	1.014	1.008	1.004	1.002	1.001	1.001	1.000			
S	2.307	2.6	1.526	1.163	1.077	1.045	1.024	1.013	1.007	1.004	1.003	1.001	1.001	1.001	1.000			
СІ	2.622	2.9	1.642	1.221	1.106	1.064	1.034	1.017	1.010	1.007	1.004	1.002	1.001	1.001	1.000			
Ar	2.957	2.5	1.507	1.154	1.074	1.045	1.024	1.013	1.008	1.004	1.002	1.001	1.001	1.000	1.000			
K	3.312	2.1	1.372	1.088	1.043	1.026	1.015	1.008	1.005	1.002	1.001	1.000	1.000	1.000	1.000			
Ca	3.690	2.1	1.358	1.080	1.039	1.024	1.013	1.008	1.005	1.001	1.000	1.000	1.000	1.000	1.000			
Sc	4.088	2.0	1.340	1.071	1.035	1.022	1.012	1.007	1.004	1.001	1.000	1.000	1.000	1.000	1.000			
Ti	4.508	2.0	1.322	1.062	1.031	1.019	1.011	1.006	1.003	1.001	1.000	1.000	1.000	1.000	1.000			
V	4.949	1.9	1.291	1.046	1.022	1.013	1.007	1.004	1.003	1.001	1.000	1.000	1.000	1.000	1.000			
Cr	5.411	1.9	1.302	1.052	1.025	1.015	1.008	1.004	1.003	1.001	1.000	1.000	1.000	1.000	1.000			
Mn	5.895	1.9	1.289	1.045	1.022	1.013	1.007	1.004	1.002	1.001	1.001	1.000	1.000	1.000	1.000			
Fe	6.400	1.9	1.286	1.044	1.021	1.013	1.007	1.005	1.003	1.001	1.000	1.000	1.000	1.000	1.000			
Со	6.925	1.9	1.288	1.045	1.022	1.013	1.007	1.004	1.003	1.001	1.000	1.000	1.000	1.000	1.000			
Ni	7.472	1.9	1.291	1.046	1.022	1.013	1.007	1.004	1.002	1.001	1.001	1.000	1.000	1.000	1.000			
Cu	8.041	1.8	1.274	1.038	1.018	1.011	1.006	1.003	1.002	1.001	1.001	1.000	1.000	1.000	1.000			
Zn	8.631	1.8	1.265	1.033	1.016	1.009	1.005	1.003	1.002	1.001	1.001	1.000	1.000	1.000	1.000			
Ga	9.243	1.8	1.274	1.038	1.018	1.011	1.006	1.003	1.002	1.001	1.001	1.000	1.000	1.000	1.000			
Ge	9.876	1.8	1.274	1.038	1.018	1.011	1.006	1.003	1.002	1.001	1.001	1.000	1.000	1.000	1.000			
As	10.532	1.8	1.274	1.038	1.018	1.011	1.006	1.003	1.002	1.001	1.001	1.000	1.000	1.000	1.000			
Se	11.210	1.8	1.274	1.038	1.018	1.011	1.006	1.003	1.002	1.001	1.001	1.000	1.000	1.000	1.000			
Br	11.907	1.9	1.284	1.042	1.020	1.012	1.006	1.003	1.002	1.001	1.001	1.000	1.000	1.000	1.000			
Pb	10.543	1.9	1.313	1.058	1.028	1.016	1.009	1.005	1.003	1.002	1.002	1.000	1.000	1.000	1.000			
		N. 1			,			4 .					0001					
		Note:	ine	value	ror M(וטטכ	Note: The value for MOUDI Stage 1 is merely an estimate, +/- 30%.											

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Appendix D

References from extensively peer reviewed USDA "Sierra Nevada Ecosystem Project" (SNEP) (1996)

Chapter 49, Air Quality in the Sierra Nevada

Thomas A. Cahill ^{1,2}, John J. Carroll ¹, Dave Campbell ², and Thomas E. Gill ^{1,2} (1) Atmospheric Sciences, Department of Land, Air and Water Resources, (2) Air Quality Group, Crocker Nuclear Laboratory, University of California, Davis, CA 95616

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Appendix E On disk

USDA "Sierra Nevada Ecosystem Project" (SNEP) (1996) Chapter 49, Air Quality in the Sierra Nevada

Thomas A. Cahill ^{1,2}, John J. Carroll ¹, Dave Campbell ², and Thomas E. Gill ^{1,2} (1) Atmospheric Sciences, Department of Land, Air and Water Resources, (2) Air Quality Group, Crocker Nuclear Laboratory, University of California, Davis, CA 95616

Appendix F On disk

USFS Lake Tahoe Watershed Assessment (2000), Chapter 3 "Air Quality", Cahill, T.A. and Cliff, S.S.

Comments Received from Professor Keith Stolzenbach

September 23, 2005

Leon Dolislager California Air Resources Board 1001 I Street PO Box 2815 Sacramento, CA 95812

Re: Review of Lake Tahoe Atmospheric Deposition Study Final Report

Dear Leon:

This letter is to transmit my review of the Final Report of the Lake Tahoe Atmospheric Deposition Study (LTADS) per the Interagency Agreement No. 98-004-66 between the University of California and the California Air Resource Board. Note that my comments reflect my professional areas of expertise. My review focuses on the inferences with regard to transport and deposition rather than on issues of air quality monitoring methodologies or atmospheric chemistry. Some of my comments below mirror and even repeat those made in my earlier reviews, but I have tried to give a fresh assessment of all issues.

General Comments

- The general approach of estimating atmospheric deposition rates by using
 observed atmospheric concentrations in conjunction with theoretical deposition
 velocities is a well-established methodology. The quantity and quality of the
 concentration and meteorological data (used as a basis for the deposition
 velocity calculation) in this study greatly exceeds that of previous studies in other
 regions.
- This report focuses entirely on direct deposition to the water surface. In a study
 of atmospheric deposition in the LA area, we found that the atmospheric loading
 to streams and water bodies was more influenced by deposition on the
 watershed that is then washed off than by direct deposition on the water surface.
 Of course, Lake Tahoe has a large surface area relative to its watershed, so this
 may be less so there.
- In my judgment, the precision of deposition estimates, whether on the basis of direct measurements or theoretical calculations, can not be considered to be better than about ±30-50%. This uncertainty is the result of limitations in measurements of temporal and spatial distribution of the atmospheric concentrations of the substances of interest as well as inadequacies of the theoretical formulations for predicting deposition.
- This study deliberately did not make much use of surrogate surfaces, correctly
 noting the issues relating to realistic collection of small particles. Here in LA we
 found that relatively simple surrogate surfaces gave us excellent estimates of dry
 deposition or particulates averaged over a season, largely because most of the

deposition was by large particles that are collected the most accurately by such surfaces. Given the results of the size dependent deposition reported in Chapter 4 (see below for my comments), such surfaces might have been very useful in LTADS.

Specific Comments by Chapter

Chapters 1-3: These chapters present mostly introductory and background material. I have the following comments:

- On page 1-12 there is a discussion of the implications of the particle size
 distribution in water. In considering this issue it should be recognized that once
 particles are deposited on the lake surface there are a variety of processes,
 including physical and biological aggregation, dissolution, and chemical
 transformation, that will make the particle size distribution in water very different
 from that in air.
- The over-water measurements presented in Chapter 3 and discussed elsewhere in the report are consistent with our findings in LA that direct deposition of urban aerosols (and probably vapor phase material) occurs particularly during diurnal periods of offshore wind. At Tahoe, where the direct deposition is the main focus of interest (as opposed to LA where we were more interested in deposition on the watershed), this is a potentially important component of the input to the lake and the LTDAS study does a very good job of documenting the temporal and spatial structure of the land-water interactions.

Chapter 4: Several of the questions I raised in my reviews of 10/4/04 and 5/31/05 regarding the theoretical basis for the deposition calculations have been addressed in the final report, either by changes in the formulation or by sensitivity analysis that shows the relative importance of particular approximations. Here I will mention only those remaining issues about which I have concern.

- Equation 4.16, representing deposition by Brownian motion and inertial impaction is quite often used, but is theoretically applicable to smooth surfaces only. This limitation is also ignored throughout the literature. There are formulations applicable to rough surfaces, and some papers correctly use them. I have not been too concerned about the error introduced into the deposition calculation because I suspect that aerosols in the size range affected by these assumptions do not contribute much to the total deposition over water. The uncertainty in the friction velocity calculation over rougher land surfaces (caused by the low measurement height relative to the roughness height) shows up dramatically in the inertial impaction term because the Stokes Number is proportional to the square of the friction velocity. In the LTADS calculations this would be important only in the near-shore region where the land-based friction velocity is used.
- I agree generally with the assessment in the report about the positive and negative biases in the calculations. This is a frank assessment and is consistent with my statements about the overall precision of deposition estimates.

- The particle size distributions reported here are consistent with our measurements in LA in that we also see a substantial fraction of the mass associated with very large (greater than 10 microns) particles that are responsible for most of the PM deposition.
- The measurements made near roadways are entirely consistent with our comparable measurements here in LA in terms of the distance downwind before the particle concentration is lowered by dispersion. This small spatial scale fits in well with the goal of understanding the spatial patterns of deposition on the lake. I would be cautious about the statement on page 4-53 that much of the material resuspended from the road deposits within a short distance. There is a substantial mass of smaller particles that do not deposit significantly and whose concentration is reduced primarily by dispersive dilution as discussed in the report.
- The temporal variations in constituent concentration and calculated deposition are interesting and are obviously dominant features at this site, largely because of the diurnal pattern of upslope and downslope air flows. However, it should be noted that the empirical methodologies used to calculate the deposition rates are based on expressions calibrated to transports averaged over at least a day. There have been few if any studies of transports at an hourly time scale. For this reason, the hourly variations in calculated deposition should be considered semiquantitative at best.
- I note in Figure 4-29 that the seasonal variation in PM and N is modest. This is
 consistent with our results in the urban areas of LA. We interpret this as partially
 the result of significant resuspension (by traffic and wind) of dust and associated
 contaminants that tend to homogenize the region and modulate both spatial and
 temporal gradients.

Chapter 5: The methods used to calculate wet deposition seem logically constructed and based on adequate data about contaminant mass in the air column, but I do not have particular expertise in this area that will allow me to identify any detailed issues with this analysis.

Chapter 6: This Chapter reviews the local, mesoscale, and regional sources of nitrogen that may be transported to Lake Tahoe and be deposited on the lake or its watershed. Much of this report deals with the atmospheric chemistry of nitrogen, which I am not qualified to evaluate. The analysis of the Sacramento urban plume is a robust combination of data analysis and modeling and the estimates of dilution are consistent with my experience with this kind of transport. It appears that the contribution of this pathway to Tahoe is limited by dilution and deposition as the air mass moves eastward.

Chapter 7: This chapter presents information about sources of nutrients and particulate matter. My comments pertain to the PM sources as I have less experience with nutrient sources.

 As in most regions, the emissions estimates, based on silt loadings and traffic densities, indicate that road dust is the major source of PM. I do not have any firm evidence to contradict this result, but our experience in LA has led us to believe that the road dust estimates may be high and windblown dust estimates may be low. One indirect piece of evidence for this is that, if the road dust emissions were as high as estimated, there is no identified mechanism for replenishment of the silt loading on the road itself. Roads can be a local source of the largest particles that are hardest to resuspend by wind, but, as shown in this report, the region of influence of these particles is confined to a 100 meters or less from the source. What we need to understand better are the true sources of dust (we think it is mostly crustal material resuspended by wind) and the dynamics of the cycle of resuspension and deposition.

• The review of aerosol data is a valuable synthesis of data, particularly with regard to the potential importance of long range transport. Our studies of deposition in Los Angeles have implicated inter-regional transport even of fairly coarse particles as a result of resuspension and transport by wind and traffic.

Chapter 8: The conclusions regarding PM sources and deposition to the lake are consistent with the analyses presented in the individual chapters.

I hope that this review is useful in your evaluation of the project final report. Please contact me with any questions you may have. In closing let me say that I intend any critical remarks to be taken constructively and that on balance I find this study to be an extremely well-done investigation of a difficult issue.

Very truly yours,

Keith D. Stolzenbach

Professor of Civil and

Environmental Engineering

Comments Received from Professor Gail Tonnesen

Appendix 1

Peer Review of "The Lake Tahoe Atmospheric Deposition Study" Final Report Submitted by Gail Tonnesen, CE-CERT, University of California Riverside

General Comments

The goal of the LTADS study was to assess the relative contributions of local and transported nutrients nitrogen (N) and phosphorus (P) and fine particulates (PM) directly deposited into Lake Tahoe. Runoff from land areas in the surrounding watershed is also a possibly significant contributor to nutrients and PM into the lake, but these sources will be assessed in a separate watershed analysis to be completed by the Lahontan Regional Water Quality Control Board and RWQCB contractors. Thus, the focus of this study is atmospheric emissions, chemical transformations, transport and direct deposition into the lake. The question of the importance of direct deposition into the lake versus run off from surrounding areas cannot be addressed until the separate watershed analysis is completed. However, it would be useful to include in the summary of this report a description of the research plan and schedule for the water shed analysis.

The contribution from runoff will include a component from atmospheric deposition and components from other land use activities, such as fertilizer use, erosion, etc. It is likely that the assessment of nutrient deposition to land and subsequent run off to the lake will make use of many of the same measurements and modeling described in this report. It would be useful to describe in this report the deposition of nutrients both directly into the lake and to land within the Lake Tahoe basin. It might also be useful to compare and contrast the Lake Tahoe Basin with the previous studies of nutrient loading in the Chesapeake Bay watershed. ARB staff are probably already familiar with the Chesapeake Bay nutrient research, but I am including here links to the USGS research: http://water.usgs.gov/wid/html/chesbay.html and to the Chesapeake Information Management System: http://www.chesapeakebay.net/info/wqcriteriapv/index.cfm#pvtoc

The ambient monitoring carried out in the LTADS study was comprehensive and included a large array of data collection that will be useful in future attempts to model the atmospheric contribution of nutrients and PM to the basin. Especially notable was the use of the collocated Beta Attenuation Monitors for refining the Two-Week-Sampler measurements and the use of particle size counters to provide size distribution data for PM.

The major focus of the LTDAS study was to estimate direct, dry and wet deposition of nutrients and PM to Lake Tahoe.. It would be interesting and useful to compare the deposition estimates from the LTDAS study to values calculated in a photochemical grid model. The LTDAS calculated deposition rates should be more accurate than grid model results because the LTDAS estimates are based on ambient measurements of species concentrations and meteorology data. Moreover, the LTDAS adopts several

detailed corrections to the deposition algorithm (e.g., corrections for the 20% of the lake surface near shores in Section 4.3.1.6). However, it would still be useful to compare the detailed, measurement based estimates of LTDAS with air quality models to see if they are generally consistent, and also to assess the possible usefulness of air quality model simulations for projecting future changes in N deposition to Lake Tahoe that may result from emissions controls both in upwind areas and locally. Because of the complex interaction of O3 photochemistry and the conversion of N between different forms, and the very large differences in deposition velocities for different N species, future changes in N deposition can not be estimated simply based on changes in emissions inventories. It will be necessary to perform air quality model simulation using ozone-VOC-NOy photochemical and aerosol transformations. Grid models are now being operated for long periods for California, including the 2002 period of the LTDAS study, and the grid model results should be compared with the LTDAS estimates. A widely use grid model, the Community Multiscale Air Quality (CMAQ) model uses the same deposition algorithms as those developed by Byun and Dennis that were referred to and used in the LTDAS approach, so the results should be comparable in some respects (with any differences resulting from errors in model simulated values or errors & missing data in the meteorology and concentration data). Annual modeling studies for calendar year 2002 are also being performed using ENVIRON Corporations CAMx model. The calendar year 2002 simulations are being funded by the Western Regional Air Partnership and included CMAQ and CAMx simulations with grid resolutions varying from 4-km to 36-km. Another advantage of using the grid model is that it will provided simulated concentrations over the lake surface, although the lack of measurements above the lake poses a problem for model validation. In any case, future studies should use a combination of both measurements and grid model simulations.

The report exhaustive in its detailed analysis of the data and the methods used to estimate dry and wet deposition. As such it represents a major accomplishment and will be the basis for future efforts to develop more accurate estimates of nutrient and PM deposition in Lake Tahoe. Finally, it would be helpful to name the individual authors who contributed to each chapter of this report, both to credit them for their work and as a point of contact for future inquiries into the work.

Detailed Comments on Chapters 1, 4 and 6 are included below.

Chapter 1 Comments

Page 1-1, last paragraph. Given the importance of agricultural and livestock operations as a source of nitrogen (N) and fine particulates, these sources should be mentioned in the list of likely sources of PM and nutrients.

Page 1-17, first paragraph: This section appears to be from the original proposal (e.g., "Three super monitoring sites are proposed..."). It should be rewritten to state what was actually implemented.

Page 1-25, Section 1.6.9: The description of the Multi-Isotope Ratio measurements is very vague. It mentions unique, new research tool based on "quantum mechanical processes" but does not provide any sort of description or references for the new method. This section should be rewritten to provide a more professional and technically complete description of this research activity.

Chapter 4 Comments:

Page 4-1, second paragraph: "This is one of several assumptions that are intended to provide a conservatively large estimate of dry deposition." The above statement implies that this study will be biased to over estimate nutrient deposition. It would be better to provide the most accurate estimate possible and also to include a more conservative, upper range estimate for use in planning purposes. The question of bias is also discussed on page 4-40, Section 4.4

Page 4-1, last paragraph. Will data collected in the LTADS study be provided to the RWQCB and its contractors for estimating dry and wet deposition to land surfaces? It might be useful to provide more details here about future interactions between the ARB and the RWQCB for integrating their studies.

Page 4-2, 2nd paragraph: "Some of the nutrients deposited over land would be assimilated before reaching the Lake." Is it possible to provide an estimate over reference to the fraction of land deposition that would not reach the lake?

Page 4-12, caption for Table 4-1: Definition needed for "crs" and "lrg".

There are several grammatical errors in Chapter 4 (two examples listed below), the report should be checked again for grammar errors.

Page 4-30, 2nd paragraph after equation 4.9: There appears to be a missing word in this sentence: "By using an iterative method it is also possible based water temperature and..." Should this be changed to: "...based *on* water temperature and..."?

Page 4-32, last sentence: "..., particularly for situations measurement heights..." should be "..., particularly for situations *with* measurement heights..."

Page 4-67, and Tables 4-12 to 4-14: The abbreviation "MT" is typically used for "Mega Tons (not Metric Tons), so the use of MT is very confusing here. (I spent quite a bit of time reviewing chapter 4 to see where the estimate of approx 150 millions tons was calculated before I went back to the exec summary and saw that it was 150 tons). It would also be helpful to include more details on the calculation on page 4-67, e.g., the Lake surface area for open water and near shore. Other ways to describe "metric tons" would be the spelling "tonnes" or long tons (as opposed to US units of 2000 ponds, spelled "ton" or "short tons").

Chapter 6 Comments

Page 6-2, first line starts: "Among nitrogen species, ammonia was presumed plentiful and ubiquitous in nature" and therefore NH3 was not targeted for study. This statement is probably incorrect. Although there are NH3 emissions from animals and it is possible that soil microbial processes are an important source of biogenic NH3, there are large uncertainties in those emissions and they are likely small in comparison to anthropogenic NH3 sources from agricultural and livestock operations in central CA. Study of the transport and fate of the anthropogenic NH3 emissions should be a key topic of the LTADS study. Because NH3 had a high deposition velocity it is likely that much of the central CA NH3 emissions deposit out (or are converted to aerosol ammonium nitrate). Moreover, the abundance of NH3 effects the gas-aerosol equilibrium of H2SO4, HNO3, NH3 and aerosol sulfate and nitrate, and NH3 can thereby affect the lifetime and transport of HNO3. The same arguments used in the report to show that transported HNO3 has a small effect on Lake Tahoe might also apply to the transport of NH3. Nonetheless, the sources, transport and fate of NH3 should be considered as part of this study.

Note that the importance of this is highlighted on page 7-3, section 7.1.1. where the authors state that" NH3 was found to be the primary component of N deposition to lake Tahoe."

Page 6-3, Reaction R4: the "prime" sign is included on "R" on the left side of the reaction but is missing on the "R" in the right side of the reaction.

Page 6-4: In the discussion of the chemistry and the list of the chemical reactions it is important to also include and discuss the hydrolysis of N2O5. Hydrolysis of N2O5 to produce 2 HNO3 by either gas phase or heterogeneous reactions is highly uncertain but is expected to be an important source of converting reactive NOx to HNO3, especially during the winter time. This could effect the lifetime and transport distance of NOy species.

Page 6-12, Section 6.7.2: There is an incomplete sentence that needs to be rewritten: "The most common peroxy nitrate, peroxy acetyl nitrate (PAN)" should be rewritten as: "The most common peroxyacyl nitrate is peroxy acetyl nitrate (PAN)."

Page 6-13, first paragraph: The text states that "almost all the precipitation falls as snow, thus wet deposition is the only sink of NOy during the winter". This statement is incorrect and might be a typo? Did the authors intend to say that there is only dry deposition? I expect that there would be both wet and dry deposition during the winter (certainly the dry deposition of HNO3 to a snow covered surface is rapid) so it would be best to delete the above statement.

Page 6-16, third line from bottom of page, the phrase "the availability to NO2 for ozone production" should be changed to "the availability *of* NO2 for ozone production"

Page 6-19, section 6.7.6: The following statement is unclear: "Figure 6-28 shows measurement of NO2 and total NOy (given as NOx at the urban locations)". I assume that NO2 is given as NOx at the urban locations because there is high NO, but that NOx is primarily as NO2 at rural sites. Might be best to state that Fig 6-28 shows "NO2 (or NOx at urban sites) and NOy."

Page 6-20, Section 6.8, Conclusions and Implications: the discussion about fire is somewhat confusing, and I'm not certain of the significance or accuracy of the statement that "Total reactive nitrogen in the region is likely at a maximum during the summer". Is this because the fires occur during the summer, or because air masses are advected away or dispersed more rapidly in the winter? Or does "the region" only refer to the Lake Tahoe area and not the urban areas? This section could be rewritten to elaborate on this and the significance of the fire emissions.

Comments Received from Professor Akula Venkatram

Lake Tahoe Atmospheric Deposition Study (LTADS)
Final Report, September 2005
Reviewed
By
Akula Venkatram
UC, Riverside

The objectives of the project summarized in this report are to 1) estimate annual dry and wet deposition of phosphorus, sulfur, nitrogen, and PM into Lake Tahoe, and 2) estimate the relative contributions of in-basin and outside basin sources to this deposition. The fulfillment of these objectives required the measurement of air quality and meteorological variables at several sites in the Lake Tahoe basin during the period November 2002 to March 2004. In addition several special studies were conducted to provide data to supplement the long-term measurements. Most of the resources of the project were spent on the observational and experimental components of the project. This has resulted in a data set that will have lasting value to the research community. Considering the limited time and effort allocated to the analysis of the data, CARB staff and their collaborators have made excellent progress towards fulfilling the objectives of the project. They have made reasonable estimates of wet and dry deposition of nitrogen, phosphorus, and PM into Lake Tahoe. They have also drawn important conclusions on the relative contributions of local and distant emissions to the deposition. The results obtained by ARB can and should be improved through further analysis of the extensive data set that has been collected. Section 8 of the report provides suggestions on further research. My comments, which are designed to complement these suggestions, address issues that are within my areas of competence.

2. Atmospheric Processes

This section provides information on wind, temperature, and precipitation patterns in the Lake Tahoe basin. Although the discussion shows how this information is relevant to the objectives of the project, most of the conclusions are based on qualitative arguments. Examples of important conclusions that require quantitative support are:

- 1. 1st paragraph, page 2-48. Materials must mix down some 700-1000 m or more to reach the Lake's surface.
- Same paragraph. The high pressure zone frequently creates temperature inversions over the land that might inhibit the vertical exchange of pollutants. However, the thermal mass of the Lake may be sufficient to induce vertical mixing......
- 3. 1st paragraph, page 2-49. Emissions originating from outside the basin will have much less opportunity to interact with the Lake.

The large number of figures on wind roses and mixed layer heights can be cut down considerably by using the information explicitly in a mathematical model. The measurements made with the RASS, radar wind profiler, and mini-sodar should be used

to quantify deposition or estimate source-receptor relationships using models that produce quantitative results.

3. Data quality and summary of ambient concentrations

Section 3.2.6 describes several experiments to examine the variation of near surface concentrations as air flows from the shore onto the lake. The major conclusion is that 'concentrations over the lake declined rapidly within a short distance of the shoreline'. Because this reduction is not accounted for explicitly in the deposition calculations, the report points out that deposition is overestimated. ARB needs to refine their calculations to demonstrate that the extensive data set that they have collected can be used to improve upon the zeroth order estimates that can be made with a much smaller set of measurements. Such refinements should rely on dispersion and deposition models to avoid the ambiguities of qualitative arguments. Let me illustrate how a simple model can provide insight into the deposition calculations.

Assume that the concentration is well mixed through a layer of thickness h_o at the shoreline before the air flows onto the lake. As the pollutant is deposited at the lake's surface, the pollutant layer will grow vertically in response to turbulence. A mass balance on this layer is given by:

$$\frac{d}{dy}(UhC) = -v_dC, \qquad (1)$$

where U is the wind speed in the pollutant layer, and v_d is the dry deposition velocity. For simplicity, assume that U does not vary with downwind distance, and that h, the vertical extent of the pollutant layer is given by:

$$h = h_o + \frac{\sigma_w x}{U}, \qquad (2)$$

where $\sigma_{\rm w}$ is the standard deviation of vertical velocity fluctuations. Substituting Equation (2) into Equation (1) and integrating yields

$$C = C_o \left[\frac{h_o U}{h_o U + \sigma_w X} \right]^{(1 + v_d / \sigma_w)}.$$
 (3)

We can show that the deposition rate, D(x), over a distance x from the shoreline is

$$\frac{D(x)}{Q} = 1 - \left[\frac{h_o U}{h_o U + \sigma_w x} \right]^{v_d / \sigma_w}, \tag{4}$$

where Q is the material flux into the lake. If v_d =0, no deposition occurs. For highly gases, such as NH₃ and HNO₃, $v_d \approx \sigma_w$, so that Equation (4) becomes

$$\frac{D(x)}{Q} = \frac{\sigma_w x}{h_o U + \sigma_w x},$$
(5)

and the relevant scale over which all the pollutant is deposited is

$$X_{d} = \frac{h_{o}U}{\sigma_{u}}.$$
 (6)

For large particles, $\,v_{_{d}}>>\sigma_{_{w}}$, and the corresponding removal scale is

$$X_{d} = \frac{h_{o}U}{v_{d}}.$$
 (7)

If we plug in some representative values for the variables in Equation (6) and (7), we find that pollutants flowing into the lake are removed completely within a few kilometers from the shoreline. This means that deposition calculations can be refined by measuring the inflow into the lake rather than making better estimates of deposition velocity. The inflow can be estimated by measuring simultaneous profiles of concentration and velocity at selected locations along the shoreline.

If we assume that deposition rate is controlled by the mass inflow into the lake, the total deposition rate over the lake is given by

$$D_{T} = 2\pi R C_{o} h_{o} U, \qquad (8)$$

if we assume a circular lake with radius R. If we compute the deposition rate using a deposition velocity over the lake, the value is

$$D_{F} = v_{d} \pi R^{2}, \qquad (9)$$

and the ratio of the 'true' to the 'false' deposition rates is

$$\frac{D_F}{D_T} = \frac{v_d R}{2Uh_o}.$$
 (10)

If we take $v_d=1$ cm/s, R=15 km, U=3 m/s, $h_o=10$ m, the ratio works out to be about 2.5. If h_o is actually 50 m, the ratio is 0.5 and the deposition might be actually underestimated using the deposition velocity method advocated by ARB. The point here is that the ratio can acquire a range of values depending on the concentration and velocity profiles in the air mass flowing onto the lake surface.

Further analysis of the data should be conducted using appropriate dispersion/deposition models, which we have shown can provide insight into the governing physics and can thus guide the design of experiments needed to quantify deposition. It is possible that ARB has all the measurements to conduct a more refined analysis.

4. Dry Atmospheric Deposition

The dry deposition rate over the lake is computed using

$$D = \int_{Lake} v_d C.dA \tag{11}$$

ARB estimated the concentration, C, from shoreline measurements, and they do point out that this might lead to overestimation. As shown earlier, this might not be true. The deposition velocity, v_d , is computed from shoreline meteorological measurements although the roughness length and temperature corresponding to the water surface is incorporated into the calculation. The actual deposition velocity is a weighted average of the shoreline value and the overwater value.

The equations used to compute atmospheric resistance assume horizontally homogenous conditions (large uniform fetches). They are not likely to apply to Lake

Tahoe, which is surrounded by trees. Calculations of deposition velocity might have to account for the fact that meteorological measurements were made below the tree canopy height; using a roughness length of 1 m does not account for the completely different nature of the flow below the canopy. The similarity equations might apply to heights of about 2.5 times the height of the canopy, and only when they incorporate the displacement height. The extensive literature on canopy flows should be consulted before the data are reanalyzed.

The similarity equations used to compute R_a are also not likely to be valid in the transition region downwind of the shoreline where most of the deposition occurs. Furthermore, the wind speed over the lake surface might be higher than that measured at the shoreline.

The deposition velocities of soluble gases and small particles are sensitive to the surface friction velocity, u_* . ARB has used the similarity wind profiles to estimate u_* . As indicated earlier, they might not apply to the Lake Tahoe situation. This explains why ARB was forced to set a minimum limit of 1/6 (s/cm) for R_a ; the estimated u_* was unrealistically high. Some of these problems could have avoided by measuring the surface shear stress directly using a sonic anemometer. If this was problem, u_* could have been inferred from measurements of vertical velocity fluctuations, σ_w . It appears that ARB does have access to turbulence data that can be used to refine calculations of atmospheric and laminar sublayer resistances, both of which are sensitive to surface friction velocity.

I have some minor comments that should be addressed if ARB chooses to revise their deposition calculations:

- 1. The comparison of R_a computed using the Byun and Dennis method with that based on Equation (4.12) is not useful because the bulk aerodynamic method does not account for stability effects.
- 2. It is preferable to rely on peer-reviewed literature rather than on the CALMET user's manual for the deposition equations.
- 3. Equation (4.8) should be first expressed in terms of the surface heat and momentum fluxes before the parameterizations for the fluxes are inserted into the definition.
- 4. Equation (4.6) has some errors: the log term should not have $(z-z_0)$, while the second term be $4.7(z-z_0)/L$. The equation should be consistent with Equation (4.10).
- 5. It is claimed in several places (first sentence of section 4.3.1.6) that the computation of R_a assumes a logarithmic wind profile. Obviously this is not true as Equation (4.6) shows; there is a logarithmic term but there are other terms that depend on z/L.
- 6. Why present Equation (4.15) if it was not used?
- 7. How were the results of section 4.5.4 (page 4-53) used in the calculations of deposition rates? Why are the conclusions relevant?

These minor comments should not distract attention from my major concerns that the similarity equations might not apply to the inhomogeneous conditions of lake Tahoe, and that using Equation (11) might lead to errors in dry deposition estimates. These issues can be settled by making some flux measurements using sonics. This data might be already available to ARB.

5. Wet Atmospheric Deposition

ARB has used available data to make first-cut estimates of wet deposition over lake Tahoe. These estimates are based on a formulation presented on the bottom of page 5-4. As far as I know, this formulation is not based on any work reported in the extensive literature on wet deposition. It might be misleading to call it a "first principles" approach. It incorporates several parameters whose values are assumed rather than determined from first principles. For example, the depth of the layer that is washed out is taken to be 3000 m for transported pollutants, and 700 m for in-basin pollutants. There is little justification for these assumptions. Furthermore, taking surface concentrations to represent average concentrations over these deep layers is a risky proposition.

The factors HW (horizontal washout fraction) and VW (vertical washout efficiency) are varied to produce a range of wet deposition estimates. Because we know almost nothing about these parameters, does this range of estimates have any meaning? The agreement with TRG bucket measurements might be accidental.

The conventional approach to estimating the wet deposition rate is based on the parameterization:

$$D_{w} = C_{a} w_{r} h p , \qquad (12)$$

where C_a is the air concentration averaged over the height of the storm, h, and w_r is the washout ratio defined as the concentration in precipitation divided by the concentration in air, and p is the precipitation amount. There is great deal of uncertainty in estimating w_r , which depends on a host of variables such as droplet size distribution, solubility of the pollutant in water, and aqueous phase chemistry etc. Seinfeld and Pandis (1998) provide details of the "first principles" approach to computing wet deposition. Estimates of wet deposition can be made with empirically derived values of w_r . However, we still need reasonable estimates of C_a and h. I would put more trust in wet deposition 'measurements' in buckets than in theoretical calculations.

6. Air Pollution Transport

This is a useful discussion of the processes that govern transport of pollutants from different sources into the Lake Tahoe basin. However, I do not see how one can estimate the transport component of the pollutant budget without some sort of mathematical model. Figures 6.6 and 6.11 present quantitative results, but they are not part of an integrated framework that a model would automatically impose on the analysis. The conclusions in section 6.8 are useful, but without some numbers attached to them it is difficult to interpret them.

7. Characterization of PM and Nutrient Sources

ARB conducted several studies to refine emission estimates of PM, N and P originating from roads, motor vehicles, and wood smoke. It is not clear whether these studies resulted in emission factors that could be used to construct a realistic emission inventory for the basin. Without such an inventory, it would be difficult to check consistency among different components of the pollutant budget for the basin. On of the conclusions from the special studies is that large fraction of the PM originates from roads. This is supported with results such as those presented in Figure 7-8. My questions are: How was the flux calculated without wind profiles? Why is the depth of

the plume from the road more than 400 m at a downwind distance of 1 m from the source?

8. Conclusions, Lessons Learned, Insights, and Recommendations

I expected to see a table that provided a pollutant budget for the basin. The components of such a budget are: Inflow, basin emissions, outflow, total wet deposition, and total dry deposition. If possible it should include dry deposition and wet deposition due to local sources to ensure consistency with local emissions.

It is clear that ARB staff recognize the uncertainties in their deposition estimates. Their suggestions to reduce these uncertainties are well thought out. I believe that if ARB does not follow up on these suggestions, the report, as it stands, will have limited value to the community.

Comments Received from Professor Anthony Wexler

LTADS Review Anthony Wexler Professor

Departments of Mechanical and Aeronautical Engineering, Civil and Environmental Engineering, and Land, Air and Water Resources, University of California, Davis, CA 95616

I will divide my comments into two categories: major and minor. Major comments concern the technical content of the report especially matters that may influence policy and future research regarding water clarity in the basin. Minor comments concern typographical and presentational matters.

Overall, this is a very impressive piece of work. The writing style is clear and to the point. With severe resource constraints, the staff did a superb job of measuring many relevant parameters and estimating deposition. The staff and associated researchers should be commended for an excellent job.

Major Comments

The CARB was charged with estimating the atmospheric contribution of nitrogen, phosphorous, and PM to Lake Tahoe. The charge is motivated by a multi-decade degradation in lake water clarity. A primary question is what inputs to the lake are contributing to this degradation, so that estimates of the atmospheric contribution will be compared to those from other sources (streambed erosion, shoreline erosion and groundwater discharge – page 4-2). By understanding which sources are the major contributors, mitigations can be implemented that restore the lake's clarity. It is this context that motivates my comments.

The staff has endeavored to provide conservative estimates of the various sources, where they define conservative as the upper bound. But is an upper bound estimate the conservative or most useful one? Since the estimates provided here will be compared with other sources, various scenarios present themselves:

- If the CARB conservative estimate is lower than the other sources, then
 conservative defined as an upper bound is the correct measure it is
 conservative because even if the answer is lower, agencies will be correct to
 direct their attention to the other sources and ignore the atmosphere as a source.
- If the CARB conservative estimate is comparable to or lower than the other sources, then conservative defined as an upper bound is not appropriate or "conservative" in that remediation measures may be applied to atmospheric sources when they may not be relevant.

I will make the case here that the LTADS conservative estimates may be an order of magnitude too high. If this is supported by the other reviewers, I recommend that the upper estimate remain as given by CARB but that the lower and best estimates be revised downward accordingly.

Dry Deposition

A key assumption for the dry deposition estimates is stated on page 4-66: "Concentrations measured near shore are assumed to be representative of both the near shore and open water areas of the lake." This is in great contrast to the data showing rapid decay in concentration near the shore (Figure 3-26, page 3-103). This is especially of concern for the large particle size factions where the background concentration may be an order of magnitude lower than the near shore value, and where the majority of the atmospheric PM resides. Using rough numbers, if we estimate the lake to be 20 km by 30 km, then the surface area is 600 km². If the particle concentrations decay to background over a distance of 60 m and if we assume that the midlake deposition is negligible because the deposition velocity is much lower than near-shore and the concentrations are much lower due to the aforementioned decay, then the effective area for deposition is 2(20km + 30km)x60m - the perimeter times the coastal boundary layer – giving an effective deposition area of only 6 km², 1/100 of the lake area estimated above. The actual area of Tahoe is more like 500 km² but this rough calculation is just done for order of magnitude illustration. The staff recognize this potential overestimate in section 4.4.1 on page 4-40.

The dry deposition estimate may not be two orders of magnitude too high because the staff's deposition velocity decreases rapidly near the coast so that mid-lake deposition values are already much lower than those near the coastline. On the other hand, nonsoluble particulates depositing near the shore may settle to the relatively shallow bottom before being transported to the deeper portions of the lake, even reducing further the effect of near shore deposition (although this is beyond the scope of the current study). The staff should use the method outlined above to re-estimate the deposition assuming this rapid decay in concentration near the coast.

Wet Deposition

Wet deposition is estimated by dividing the season into times of regional and local contributions. The argument that the local contribution may be too high is the same as for the dry deposition – using near shore ambient concentrations to represent those over the whole lake is probably an over estimate by an order of magnitude or so. The argument for the regional contribution is that during frontal passages, the Big Hill concentrations of pollutions should be many orders of magnitude lower than during more stagnant events without precipitation. For the regional wet deposition contribution, staff used the average concentrations at Big Hill but during frontal passages, these concentrations should be an order of magnitude or more lower than the average again leading to an over estimate by a factor of 10 or so.

Remarkably, the staff estimates of wet deposition are very close to those estimated by TRG and NADP (Table 5-11 on page 5-25). The staff express reservations about the TRG sampler at the Wallis Residence, but otherwise no information is given about possible uncertainties in these measurements. Due to the agreement between the wet deposition calculations and the TRG/NADP estimates, I have lower confidence in my contention that the upper estimates are an order of magnitude larger than the best estimate. More information about the TRG and NADP estimates might help clarify.

Minor

What is the possible contribution from wildfires – not addressed well in the study because wildfires did not occur during the measurements?

Pg. 1-16 – no start on the Big Hill site

Pg 1-22, section 1.6.3 – define RWC

Pg 1-26, section 1.6.11 – sentence should read "... proximity to major pollutant sources."

Pg 3-51, para 2 – why was 1.2 used?

Pg 4-57, line 2 – remove the hanging "c"

Pg 4-59, last para – change figure number from 4-24 to 4-26 and 4-23 to 4-25

Pg 4-72, line 4 – should read "dominated by contributions"

Pg 4-72, para 2, line 1 – change 4-27 to 4-29

Pg 4-72, para 3, line 5 – should read "heavily traveled road"